(19)【発行国】日本国特許庁（ＪＰ）

(12)【公報種別】公開特許公報（Ａ）

(11)【公開番号】特開平９－１９９１７５

(43)【公開日】平成９年（１９９７）７月３１日

(54)【発明の名称】ポリマー電解質二次電池の製造方法

(51)【国際特許分類第６版】

H01M 10/40

2/08

4/02

4/74

【ＦＩ】

H01M 10/40 Z

2/08 K

4/02 C

4/74 C

【審査請求】未請求

【請求項の数】５

【出願形態】ＯＬ

【全頁数】１４

(21)【出願番号】特願平８－９４２７

(22)【出願日】平成８年（１９９６）１月２３日

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(19) [Publication Office] Japanese Patent Office (JP)

(12) [Kind of Document] Japan Unexamined Patent Publication (A)

(11) [Publication Number of Unexamined Application] Japan Unexamined Patent Publication Hei 9 - 199175

(43) [Publication Date of Unexamined Application] 1997 (1997) July 31 day

(54) [Title of Invention] MANUFACTURING METHOD OF POLYMER ELECTROLYTE SECONDARY BATTERY

(51) [International Patent Classification 6th Edition]

H01M 10/40

2/08

4/02

4/74

[FI]

H01M 10/40 Z

2/08 K

4/02 C

4/74 C

[Request for Examination] Examination not requested

[Number of Claims] 5

[Form of Application] OL

[Number of Pages in Document] 14

(21) [Application Number] Japan Patent Application Hei 8 - 9427

(22) [Application Date] 1996 (1996) January 23 days

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(57)【要約】

【課題】　外装部材とこの中に収納された素電池との間の空隙量を著しく低減することが可能なポリマー電解質二次電池の製造方法を提供する。

【解決手段】　ポリマー電解質素電池を熱融着性樹脂面を持つ外装フィルムで収納した構造のポリマー電解質二次電池を製造する方法において、第１長尺フィルムの熱融着性樹脂面上に複数の前記素電池を前記フィルムの長さ方向に所望の間隔をあけて設置し、第２長尺フィルムの熱融着性樹脂面を複数の前記素電池上面を覆うように重ね、前記第２長尺フィルム上に厚さが前記素電池と同等もしくは僅かに厚いガイド板を少なくとも前記素電池の前記フィルムの長さ方向に沿う側壁に近接するように配置し、前記２枚の長尺フィルムを加熱された一対の弾性ロールの間に通過させて前記各素電池周辺の２枚の長尺フィルムを加熱加圧して熱シールする工程とを具備したことを特徴とする。

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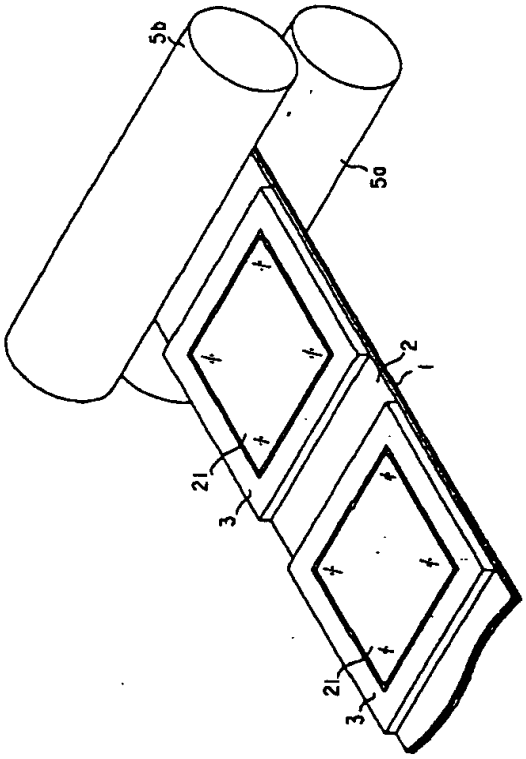
(57) [Abstract]

[Problem]　Manufacturing method of polymer electrolyte secondary battery whose it is possible to decrease void amount withthe externally mounted member and unit battery which among these is stored up considerably isoffered.

[Means of Solution]　Polymer electrolyte unit battery in method which produces polymer electrolyte secondary battery of structure which isstored up with outdoor film which has hot melt adhesion resin surface regarding, Opening desired interval to longitudinal direction of aforementioned film, itinstalls aforementioned unit battery of plural on hot melt adhesion resin surface of the 1st lengthwise film, In order hot melt adhesion resin surface of 2nd lengthwise film to cover aforementioned unit battery upper surface ofthe plural, to pile up, In order guide plate where thickness is thick equally to theaforementioned unit battery or barely on aforementioned 2nd lengthwise film proximityto do in side wall which at least parallels to longitudinal direction of theaforementioned film of aforementioned unit battery, it arranges, Is heated passing aforementioned 2 lengthwise film between elastic roll of thepair which heating and pressurizing doing 2 lengthwise film of aforementioned eachunit battery periphery, it designates that

step which hot seal is done ispossessed as feature.



【特許請求の範囲】|

【請求項１】　集電体に活物質および非水電解液を保持するポリマーを含む正極層を積層した正極と、集電体にリチウムイオンを吸蔵放出する炭素質材料を含む負極層を積層した負極と、前記正極の正極層および前記負極の負極層の間に介在された非水電解液を保持するポリマーを含む固体ポリマー電解質層とを備えたポリマー電解質素電池を外装部材で収納した構造のポリマー電解質二次電池を製造する方法において、

第１長尺フィルムの熱融着性樹脂面上に複数の前記素電池を前記フィルムの長さ方向に所望の間隔をあけて設置する工程と、第２長尺フィルムの熱融着性樹脂面を複数の前記素電池上面を覆うように重ねる工程と、前記第２長尺フィルム上に厚さが前記素電池と同等もしくは僅かに厚いガイド板を少なくとも前記各素電池の前記フィルムの長さ方向に沿う側壁に近接するようにそれぞれ配置し、前記ガイド板の底面で前記２枚の長尺フィルムを互いに重ねる工程と、前記複数の素電池を間に挟んだ２枚の長尺フィルムを加熱された一対の耐熱

[Claim(s)]

[Claim 1]　Laminates positive electrode layer which includes p lymer which keeps active substance andthe nonaqueous electrolyte solution in current collector positive electrode whicl Regarding to method which produces polymer electrolyte secondary battery of structure whichstores up polymer electrolyte unit battery which has with negative electrode whicl laminates negative electrode layerwhich includes carbonaceous material which lithium ion intercalation and release is done and thesolid polymer electroiyte layer which includes polymer which keeps positive electrode layer of theaforementioned positive electrode and nonaqueous electrolyte solution which lies between between thenegative electrode layer of aforementioned negative electrode with externally mounted member in current collector,

Step which opening desired interval to longitudinal direction of aforementionedfilm, installs aforementioned unit battery of plural on hot melt adhesion resin surface ofthe 1st lengthwise film. step which in order to cover aforementioned unit battery upper surface of theplural, repeats hot melt adhesion resin surface of 2nd lengthwise film. In order guide plate where thickness is thick equally to theaforementioned unit battery or barely on aforementioned 2nd lengthwise film proximityto do in side wall which at least parallels to longitudinal direction of

性の弾性ロールの間に通過させて前記各素電池周辺の前記２枚の長尺フィルムを加熱加圧してそれらの熱融着樹脂面を熱シールする工程とを具備したことを特徴とするポリマー電解質二次電池の製造方法。

theaforementioned film of aforementioned each unit battery, step whichit arranges respectively, repeats aforementioned 2 lengthwise film mutuallywith bottom surface of aforementioned guide plate. Is heated passing 2 lengthwise film which put between unit battery of theaforementioned plural between between elastic roll of heat resistance of thepair which heating and pressurizing doing aforementioned 2 lengthwise film of theaforementioned each unit battery periphery, manufacturing method of polymer electrolyte secondary battery which designatesthat step which those hot melt adhesion resin surface hot seal is done is possessed asfeature.

【請求項２】　前記正極および負極に用いられる集電体のいずれ一方もしくは両者は、前記非水電解液が流通可能な構造を有することを特徴とする請求項１記載のポリマー電解質二次電池の製造方法。|

[Claim 2] Anyway one side of current collector which is used : or aforementionedpositive electrode and negative electrode or as for both, aforementioned nonaqueous electrolyte solution manufacturing method of thepolymer electrolyte secondary battery which is stated in Claim 1 which designates that it possessesthe structure which is circulation possible as feature.

【請求項３】　集電体に活物質および非水電解液を保持するポリマーを含む正極層を積層した正極と、集電体にリチウムイオンを吸蔵放出する炭素質材料を含む負極層を積層した負極と、前記正極の正極層および前記負極の負極層の間に介在された非水電解液を保持するポリマーを含む固体ポリマー電解質層とを備えたポリマー電解質素電池を外装部材で収納した構造のポリマー電解質二次電池を製造する方法において、

[Claim 3] Laminates positive electrode layer which includes p· lymer which keeps active substance andthe nonaqueous electrolyte solution in current collector positive electrode whicl Regarding to method which produces polymer electrolyte secondary battery of structure whichstores up polymer electrolyte unit battery which has with negative electrode whicl laminates negative electrode layerwhich includes carbonaceous material which lithium ion intercalation and release is done and thesolid polymer electrolyte layer which includes polymer which keeps positive electrode layer of theaforementioned positive electrode and nonaqueous electrolyte solution which lies between between thenegative electrode layer of aforementioned negative electrode with externally mounted member in current collector,

第１長尺フィルムの熱融着性樹脂面上に複数の前記素電池を前記フィルムの長さ方向に所望の間隔をあけて設置する工程と、前記長尺フィルム上に厚さが前記素電池と同等もしくは僅かに厚い高分子樹脂からなるスペーサを少なくとも前記各素電池の前記フィルムの長さ方向に沿う側壁に接触するようにそれぞれ設置する工程と、第２長尺フィルムの熱融着性樹脂面を複数の前記素電池および前記スペーサの上面を覆うように重ねる工程と、前記複数の素電池およびスペーサを間に挟んだ２枚の長尺フィルムを加熱された一対の耐熱性の弾性ロールの間に通過させて前記各素電池周辺の前記２枚の長尺フィルムを少なくとも前記スペーサを挟んで加熱加圧して熱シールする工程とを具備したことを特徴とするポリマー電解質二次電池の製造方法。

Step which opening desired interval to longitudinal direction of aforementionedfilm, installs aforementioned unit battery of plural on hot melt adhesion resin surface ofthe 1st lengthwise film. step which in order to contact side wall which at least parallelsto longitudinal direction of aforementioned film of aforementioned eachunit battery, installs spacer which consists of polymer resin where thicknessis thick equally to aforementioned unit battery or barely on theaforementioned lengthwise film respectively. step which in order to cover uppe surface of aforementionedunit battery and aforementioned spacer of plural, repeats hot melt adhesion resin surface ofthe 2nd lengthwise film. Is heated passing unit battery of aforementioned plural and 2 lengthwise filmwhich put between spacer between between elastic roll of heat resistance ofthe pair which manufacturing method of polymer electrolyte secondary battery which designates that itpossesses step which putting between aforementioned spacer atleast, heating and pressurizing doing, hot seal does aforementioned 2 lengthwise film of theaforementioned each unit battery periphery as feature.

【請求項４】　前記正極および負極に用いられる集電体のいずれ一方もしくは両者は、前記非水電解液が流通可能な構造を有することを特徴とする請求項３記載のポリマー電解質二次電池の製造方法。|

[Claim 4] Anyway one side of current collector which is used : or aforementionedpositive electrode and negative electrode or as for both, aforementioned nonaqueous electrolyte solution manufacturing method of thepolymer electrolyte secondary battery which is stated in Claim 3 which designates that it possessesthe structure which is circulation possible as feature.

【請求項５】　前記スペーサは、熱融着性樹脂からなることを特徴とする請求項３記載のポリマー電解質二次電池の製造方法。

[Claim 5] As for aforementioned spacer, manufacturing metho d of polymer electrolyte secondary battery which isstated in Claim 3 which designates that it consists of hot melt adhesion resin asfeature.

【発明の詳細な説明】

[Description of the Invention]

【０００１】

[0001]

【発明の属する技術分野】本発明は、固体ポリマー電解質層を備えるポリマー電解質二次電池に関するものである。

[Technological Field of Invention] This invention is something regarding polymer electrolyte secondary battery which has solid polymer electrolyte layer.

【０００２】

[0002]

【従来の技術】近年、電子機器の発達にともない、小型で軽量、かつエネルギー密度が高く、更に繰り返し充放電が可能な二次電池の開発が要望されている。このような二次電池としては、リチウムまたはリチウム合金を活物質とする負極と、モリブデン、バナジウム、チタンあるいはニオブなどの酸化物、硫化物もしくはセレン化物を活物質とする正極とを具備したリチウム二次電池が知られている。しかしながら、リチウムまたはリチウム合金を活物質とする負極を備えた二次電池は、充放電サイクルを繰り返すと負極にリチウムのデンドライトが発生するため、充放電サイクル寿命が短いという問題点がある。

[Prior Art] Recently, attendant upon advancement of electroni c equipment, light weight , atthe same time energy density is high with miniature, furthermoredevelopment of secondary battery where repetitive charge-discharge is possible is demand As this kind of secondary battery, lithium secondary battery which possesses with positive electrode whichdesignates negative electrode and molybdenum , vanadium , titanium or niobium or other oxide ,the sulfide or selenide which designate lithium or lithium alloy as active substance as theactive substance is known. But, secondary battery which has negative electrode which designates lithium or lithium alloy as theactive substance, when charge-discharge cycle is repeated, because dendrite of lithiumoccurs in negative electrode, is a problem that charge-discharge cycle lifetime is short.

【０００３】このようなことから、負極に、例えばコークス、黒鉛、炭素繊維、樹脂焼成体、熱分解気相炭素のようなリチウムイオンを吸蔵放出する炭素質材料を用い、$LiPF_6$のような電解質およびエチレンカーボネート、プロピレンカーボネートのような非水溶媒からなる電解液を用いた非水溶媒二次電池が提案されている。前記非水溶媒二次電池は、デンドライト析出による負極特性の劣化を改善することができるため、電池寿命と安全性を向上することができる。

[0003] From this kind of thing, nonaqueous solvent secondary battery which uses electrolyte solution which consistsof nonaqueous solvent like electrolyte and ethylene carbonate and propylene carbonate like LiPF6 making use of carbonaceous material which lithium ion like for example coke , graphite , the carbon fiber , resin pyrolysate and thermally decomposed carbon vapor intercalation and release is done, is proposed to the negative electrode. Because deterioration of negative electrode characteristic with dendrite deposition can be improved, the battery lifetime and safety it can improve aforementioned nonaqueous solvent secondary battery.

【０００４】一方、米国特許第５，２９６，３１８号明細書には正極、負極および電解質層にポリマーを添加することにより柔軟性が付与されたハイブリッドポリマー電解質を有す

[0004] Rechargeable lithium intercalation battery which posses ses hybrid polymer electrolyte where on one hand, flexibility isgranted by adding polymer to positive electrode , negative

る再充電可能なリチウムインターカレーション電池、つまりポリマー電解質二次電池が開示されている。このようなポリマー電解質二次電池は、集電体に活物質、非水電解液およびこの電解液を保持するポリマーを含む正極層を積層した正極と集電体にリチウムイオンを吸蔵放出し得る炭素質材料、非水電解液およびこの電解液を保持するポリマーを含む負極層を積層した負極との間に非水電解液およびこの電解液を保持するポリマーを含む固体ポリマー電解質層が介在された構造の素電池を備え、この素電池を外装部材内に収納した構造を有する。|

electrode and electrolyte layer inthe U. S. Patent No. 5,296, 318 specification , in other words polymer electrolyte secondary battery is disclosed. As for this kind of polymer electrolyte secondary battery, In current collector active substance , intercalation and release it can do lithium ion in positive electrode and current collector whichlaminate positive electrode layer which includes polymer which keeps nonaqueous electrolyte solution andthis electrolyte solution carbonaceous material , It has unit battery of structure where solid polymer electrolyte layer which includes polymerwhich keeps nonaqueous electrolyte solution and this electrolyte solution with negative electrode which laminatesthe negative electrode layer which includes polymer which keeps nonaqueous electrolyte solution and thiselectrolyte solution lies between, it possesses structure which stores up this unit batteryinside externally mounted member.

【０００５】ところで、前記素電池を前記外装部材内に収納するには、従来、例えばポリエチレンのような熱融着性フィルムから一辺が開口された扁平状の未密封袋を作製し、未密封袋内に前記素電池を挿入し、前記開口部を熱シールして前記素電池を密封袋（外装部材）に収納することが行われている。しかしながら、このような方法で製造されたポリマー電解質二次電池は前記未密封袋内への前記素電池の挿入、開口部の熱シールにおいて十分な脱気がなされないために袋状の外装部材と素電池の間に高容量の空隙を生じ、前記素電池からの非水電解液の流出による電池性能を低下を招く問題があった。

[0005] By way, aforementioned unit battery is stored up inside theaforementioned externally mounted member, not yet sealed bag of flat where untilrecently, one edge is opened from hot melt adhesion film like for example polyethylene isproduced, aforementioned unit battery is inserted into not yet sealed bag, the aforementioned opening part hot seal is done and storing up theaforementioned unit battery in sealed bag (externally mounted member) is done. But, as for polymer electrolyte secondary battery which is produced with this kind of methodbecause you cannot do sufficient outgassing in hot seal of insertion and the opening part of aforementioned unit battery to inside aforementioned not yetsealed bag to cause gap of high capacity in externally mounted member of bag andbetween unit battery, battery performance due to outflow of nonaqueous electrolyte solution from theaforementioned unit battery there was a problem which causes decrease.

【０００６】すなわち、ポリマー電解質二次電池は充放電時に前記素電池を構成する正極層、負極層および固体ポリマー電解質層が膨脹・収縮を繰り返すため、それらの中に保持された非水電解液が前記素電池から前記外装部材内に滲み出し易くなる。特に、後述する非水電解液の素電池への供給のために前記素電池を構成する正極および負極の集電体の少なくとも一方がパンチドメタルのような非水電解液が流通し得る構造を有すると、前記充放電時に前記素電池内の非水電解液が前記パンチドメタルからなる集電体を通して前記外装部材内に滲み出し易くなる。従来のポリマー電解質二次電池は前記外装部材と前記素電池の間に高容量の空隙が存在し、前記二次電池の充放電時における前記素電池を構成する正極層、負極層および固体ポリマー電解質層が膨脹を抑制する作用が劣るため、前記素電池からの非水電解液の滲み出しが生じる。また、滲み出した非水電解液は前記空隙に溜まる、つまり素電池内の非水電解液量が低下する。その結果、充放電の繰り返しにより電池容量が急激に低下する問題があった。

[0006] As for namely, polymer electrolyte secondary battery because positive electrode layer , negative electrode layer and solid polymer electrolyte layer which formthe aforementioned unit battery at time of charge-discharge repeat expansion *contraction, nonaqueous electrolyte solution which is kept among those from theaforementioned unit battery becomes exuding easy inside aforementionedexternally mounted member Especially, when it possesses structure to which nonaqueous electrolyte solution at least oneof current collector of positive electrode and negative electrode which forms aforementioned unit battery forsupplying to unit battery of nonaqueous electrolyte solution which it mentions later like thepunched metal can circulate, it becomes exuding easy inside theaforementioned externally mounted member through current collector where nonaqueous electrolyte solution inside theaforementioned unit battery consists of aforementioned punched metal at timeof aforementioned charge-discharge. As for conventional polymer electrolyte secondary battery gap of high capacity exists between theaforementioned externally

mounted member and aforementioned unit battery, because actionwhere positive electrode layer , negative electrode layer and solid polymer electrolyte layer which form aforementionedunit battery at time of charge-discharge of aforementioned secondary battery control theexpansion is inferior, exuding of nonaqueous electrolyte solution from aforementionedunit battery occurs. In addition, exuding is nonaqueous electrolyte solution accumulates in aforementionedgap, in other words amount of nonaqueous electrolyte solution inside unit battery decreases. As a result, there was a problem where battery capacity decreases suddenly withthe repetition of charge-discharge.

【０００７】

[0007]

【発明が解決しようとする課題】本発明は、外装部材とこの中に収納された素電池との間の空隙量を著しく低減することが可能なポリマー電解質二次電池の製造方法を提供しようとするものである。

[Problems to be Solved by the Invention] This invention is som ething which it tries to offer manufacturing method of polymer electrolyte secondary batterywhose it is possible to decrease void amount with externally mounted member and theunit battery which among these is stored up considerably.

【０００８】本発明は、少なくとも平行する２つの側面にスペーサを有する外装部材とこの中に収納された素電池との間の空隙量を著しく低減することが可能なポリマー電解質二次電池の製造方法を提供しようとするものである。

[0008] This invention is something which it tries to offer manu facturing method of polymer electrolyte secondary batterywhose it is possible to decrease void amount with externally mounted member whichpossesses spacer in 2 side face which is parallel at least and theunit battery which among these is stored up considerably.

【０００９】

[0009]

【課題を解決するための手段】本発明に係るポリマー電解質二次電池は、集電体に活物質および非水電解液を保持するポリマーを含む正極層を積層した正極と、集電体にリチウムイオンを吸蔵放出する炭素質材料を含む負極層を積層した負極と、前記正極の正極層および前記負極の負極層の間に介在された非水電解液を保持するポリマーを含む固体ポリマー電解質層とを備えたポリマー電解質素電池を外装部材で収納した構造のポリマー電解質二次電池を製造する方法において、第１長尺フィルムの熱融着性樹脂面上に複数の前記素電池を前記フィルムの長さ方向に所望の間隔をあけて設置する工程と、第２長尺フィルムの熱融着性樹脂面を複数の前記素電池上面を覆うように重ねる工程と、前記第２長尺フィルム上に厚さが前記素電池と同等もしくは僅かに厚いガイド板を少なくとも前記各素電池の前記フィルムの長さ方向に沿う側壁に近接するようにそれぞれ配置し、前記ガイド板の底面で前記２枚の長尺フィルムを互いに重ねる工程と、前記複数の素電池を前記ガイド板により間に挟んだ２枚の長尺フィルムを加熱された一対の耐熱性の弾性ロールの間に通過させて前記各素電池周辺の前記２枚の長尺フィルムを加熱加圧してそれらの熱融着樹脂面を熱シールする工程とを具備したことを特徴とするものである。

[Means to Solve the Problems] Relates to this invention as for polymer electrolyte secondary battery which, Laminates positive electrode layer which includes polymer which keeps active substance andthe nonaqueous electrolyte solution in current collector positive electrode which, In current collector laminates negative electrode layer which includes carbonaceous material which thelithium ion intercalation and release is done negative electrode which, Regarding to method which produces polymer electrolyte secondary battery of structure whichstores up polymer electrolyte unit battery which has with solid polymer electrolyte layer which includes polymerwhich keeps positive electrode layer of aforementioned positive electrode and nonaqueous electrolyte solution whichlies between betweer negative electrode layer of aforementioned negative electrode with theexternally mounted member, step which opening desired interval to longitudinal direction of theaforementioned film, installs aforementioned unit battery of pluralon hot melt adhesion resin surface of 1st lengthwise film. step which in order to cover aforementioned unit battery upper surface of theplural, repeats hot melt adhesion resin surface of 2nd lengthwise film. In order guide plate where thickness is thick equally to theaforementioned unit battery or barely on aforementioned 2nd lengthwise film proximityto do in side wall

which at least parallels to longitudinal direction of theaforementioned film of aforementioned each unit battery, step whichit arranges respectively, repeats aforementioned 2 lengthwise film mutuallywith bottom surface of aforementioned guide plate. unit battery of aforementioned plural is heated passing 2 lengthwise filmwhich put between between with aforementioned guide plate between theelastic roll of heat resistance of pair which heating and pressurizing doing theaforementioned 2 lengthwise film of aforementioned each unit battery periphery, it is somethingwhich designates that step which those hot melt adhesion resin surface hot seal is done ispossessed as feature.

【００１０】本発明に係わる別のポリマー電解質二次電池は、集電体に活物質および非水電解液を保持するポリマーを含む正極層を積層した正極と、集電体にリチウムイオンを吸蔵放出する炭素質材料を含む負極層を積層した負極と、前記正極の正極層および前記負極の負極層の間に介在された非水電解液を保持するポリマーを含む固体ポリマー電解質層とを備えたポリマー電解質素電池を外装部材で収納した構造のポリマー電解質二次電池を製造する方法において、第１長尺フィルムの熱融着性樹脂面上に複数の前記素電池を前記フィルムの長さ方向に所望の間隔をあけて設置する工程と、前記長尺フィルム上に厚さが前記素電池と同等もしくは僅かに厚い高分子樹脂からなるスペーサを少なくとも前記各素電池の前記フィルムの長さ方向に沿う側壁に接触するようにそれぞれ設置する工程と、第２長尺フィルムの熱融着性樹脂面を複数の前記素電池および前記スペーサの上面を覆うように重ねる工程と、前記複数の素電池およびスペーサを間に挟んだ２枚の長尺フィルムを加熱された一対の耐熱性の弾性ロールの間に通過させて前記各素電池周辺の前記２枚の長尺フィルムを少なくとも前記スペーサを挟んで加熱加圧して熱シールする工程とを具備したことを特徴とするものである。

[0010] Relates to this invention as for another polymer electro lyte secondary battery which, Laminates positive electrode layer which includes polymer which keeps active substance andthe nonaqueous electrolyte solution in current collector positive electrode which, In current collector laminates negative electrode layer which includes carbonaceous material which thelithium ion intercalation and release is done negative electrode which, Regarding to method which produces polymer electrolyte secondary battery of structure whichstores up polymer electrolyte unit battery which has with solid polymer electrolyte layer which includes polymerwhich keeps positive electrode layer of aforementioned positive electrode and nonaqueous electrolyte solution whichlies between between negative electrode layer of aforementioned negative electrode with theexternally mounted member, step which opening desired interval to longitudinal direction of theaforementioned film, installs aforementioned unit battery of pluralon hot melt adhesion resin surface of 1st lengthwise film. step which in order to contact side wall which at least parallelsto longitudinal direction of aforementioned film of aforementioned eachunit battery, installs spacer which consists of polymer resin where thicknessis thick equally to aforementioned unit battery or barely on theaforementioned lengthwise film respectively, step which in order to cover upper surface of aforementionedunit battery and aforementioned spacer of plural, repeats hot melt adhesion resin surface ofthe 2nd lengthwise film. Is heated passing unit battery of aforementioned plural and 2 lengthwise filmwhich put between spacer between between elastic roll of heat resistance ofthe pair which it is something which designate that it possesses thestep which putting between aforementioned spacer at least, theheating and pressurizing doing, hot seal does aforementioned 2 lengthwise film of theaforementioned each unit battery periphery as feature.

【００１１】

【発明の実施の形態】以下、本発明に係るポリマー電解質二次電池の製造方法を図１～図４を参照して詳細に説明する。まず、図１に示すように第１長尺フィルム１の熱融着性樹脂面上に複数の例えば矩形状のポリマー電解質素電池２１を前

[0011]

[Embodiment of Invention] Below, referring to Figure 1 to Figu re 4, you explain manufacturing method of polymer electrolyte secondary batterywhich relates to this invention in detail. First as shown in Figure 1, opening desired interval to longitudinal

記フィルム１の長さ方向に所望の間隔をあけて設置する。前記各素電池２１を前記第１長尺フィルム１に設置するにあたっては、接着剤を用いて前記長尺フィルム１に仮固定することが好ましい。前記素電池２１は、図２に示すように正極２２と負極２３との間に固体ポリマー電解質層２４が介装された構造を有する。前記正極２２は、正極集電体２５と、この集電体２５に担持された正極層２６から構成されている。前記負極２３は、負極集電体２７と、この集電体２７に担持された負極層２８とから構成されている。

direction of theaforementioned film 1, it installs polymer electrolyte unit battery 21 of for example rectangle of pluralon hot melt adhesion resin surface of 1st lengthwise film 1. When aforementioned each unit battery 21 is installed in aforementioned1st lengthwise film 1, to do it is desirable temporary affixion to do in aforementionedlengthwise film 1 making use of adhesive and. Aforementioned unit battery 21, as shown in Figure 2, has structure wherethe solid polymer electrolyte layer 24 is introduced with positive electrode 22 and negative electrode 23. Aforementioned positive electrode 22 is formed from positive electrode layer 26 which is borne inthe positive electrode collector 25 and this current collector 25. Aforementioned negative electrode 23 is formed from negative electrode layer 28 which is borne inthe negative electrode collector 27 and this current collector 27.

【００１２】次いで、同図１に示すように第２長尺フィルム２の熱融着性樹脂面を複数の前記素電池２１上面を覆うように重ねた後、前記第２長尺フィルム２上に厚さが前記素電池２１と同等もしくは僅かに厚い枠状のガイド板３を前記各素電池２１の周囲側壁に近接するようにそれぞれ配置し、前記ガイド板３の底面で前記２枚の長尺フィルム１、２を互いに重ねる。つづいて、前記複数の素電池２１が前記ガイド板３により間に挟まれた２枚の長尺フィルム１、２を軸４ａ、４ｂを有する耐熱性の弾性ロール５ａ、５ｂの間に通過させる。前記ゴムロール５ａ、５ｂは、長さが前記長尺フィルム１、２の幅よりも十分に長く、かつ所望の加熱源により例えば前記熱融着性樹脂の軟化点以上に加熱されている。図３に示すように前記２枚の長尺フィルム１、２を加熱された前記弾性ロール５ａ、５ｂの間に通過させ、前記素電池２１および前記ガイド板３が前記弾性ロール５ａ、５ｂの間に移動すると、前記弾性ロール５ａ、５ｂが前記素電池２１および前記ガイド板３を包み込むように変形し、前記ガイド板３の底面で互いに重ねられた前記２枚の長尺フィルム１、２において下面側の長尺フィルム１部分は前記下部側の弾性ロール５ａからの熱を受け、上面側の長尺フィルム２部分は前記上部側の弾性ロール５ｂから前記枠状のガイド板３を通して熱を受けて、それらの熱融着性樹脂面同士が熱シールされる。その結果、前記弾性ロール５ａ、５ｂ間を通過させた２枚の長尺フィルム１、２から前記ガイド板３を外し、前記２枚の長尺フィルム１、２を前記ガイド板３周縁が位置された箇所に沿って切断することにより図４に示すように素電池２１とこの素電池２１を収納し、その周囲側壁付近に位置した枠状の熱シール部６を有する熱融着性樹脂面を持つ２枚のフィルム７ａ、７ｂにより形成された外装部材８とから構成されたポリマー電解質二次電池９が製造される。

[0012] Next, way it shows in same Figure 1, in order hot melt adhesion resin surface of 2nd lengthwise film 2 tocover aforementioned unit battery 21 upper surface of plural, after repeating, in orderthe guide plate 3 of frame where thickness is thick equally to theaforementioned unit battery 21 or barely on aforementioned 2nd lengthwise film 2 proximityto do in periphery side wall of aforementioned each unit battery 21, it arrangesrespectively, repeats aforementioned 2 lengthwise film 1, 2 mutually with bottom surfaceof aforementioned guide plate 3. Continuing, unit battery 21 of aforementioned plural it passes 2 lengthwise film 1, 2which was put between between by aforementioned guide plate 3 between theelastic roll 5a, 5b of heat resistance which possesses axial 4a, 4b. Aforementioned rubber roll 5a, 5b length is long in fully in comparisonwith width of aforementioned lengthwise film 1, 2, is heated to softening point or higher ofthe for example aforementioned hot melt adhesion resin at same time by desired heat source . As shown in Figure 3, is heated passing aforementioned 2 lengthwise film 1, 2between aforementioned elastic roll 5a, 5b which, Aforementioned unit battery 21 and aforementioned guide plate 3 move between theaforementioned elastic roll 5a, 5b when, In order for aforementioned elastic roll 5a, 5b to wrap aforementioned unit battery 21and aforementioned guide plate 3, it becomes deformed, lengthwise film 1 portion of under side receives heat from elastic roll 5a of theaforementioned bottom side in aforementioned 2 lengthwise film 1, 2 which is repeatedmutually with bottom surface of aforementioned guide plate 3, as for lengthwise film two portionsof top side receiving heat through guide plate 3 of aforementionedframe from elastic roll 5b of aforementioned upper side, those hot melt adhesion resin surface are donethe hot seal. As a result, From 2 lengthwise film 1, 2 which passes between aforementioned elastic roll 5a, 5b theaforementioned guide plate 3 to remove, As shown in Figure 4 by cutting off aforementioned 2 lengthwise film 1, 2alongside site where aforementioned guide plate 3 peripheral edge is located, theunit battery 21 and this unit battery 21 are stored up, polymer electrolyte secondary

【００１３】次に、素電池２１を構成する固体ポリマー電解質層２４、正極集電体２５、正極層２６、負極集電体２７および負極層２８と、長尺フィルム１、２、ガイド板３および弾性ロール５ａ、５ｂについて説明する。

【００１４】１）固体ポリマー電解質層２４

このポリマー電解質層２４は非水電解液及びこの電解液を保持するポリマーを含む。

【００１５】前記非水電解液は、非水溶媒に電解質を溶解することにより調製される。前記非水溶媒としては、エチレンカーボネート（ＥＣ）、プロピレンカーボネート（ＰＣ）、ブチレンカーボネート（ＢＣ）、ジメチルカーボネート（ＤＭＣ）、ジエチルカーボネート（ＤＥＣ）、エチルメチルカーボネート（ＥＭＣ）、γ－ブチロラクトン（γ－ＢＬ）、スルホラン、アセトニトリル、１，２－ジメトキシエタン、１，３－ジメトキシプロパン、ジメチルエーテル、テトラヒドロフラン（ＴＨＦ）、２－メチルテトラヒドロフラン等を挙げることができる。前記非水溶媒は、単独で使用しても、２種以上混合して使用しても良い。

【００１６】前記電解質としては、例えば、過塩素酸リチウム（$LiClO_4$）、六フッ化リン酸リチウム（$LiPF_6$）、ホウ四フッ化リチウム（$LiBF_4$）、六フッ化砒素リチウム（$LiAsF_6$）、トリフルオロメタンスルホン酸リチウム（$LiCF_3SO_3$）、ビストリフルオロメチルスルホニルイミドリチウム［$LiN(CF_3SO_3)_2$］等のリチウム塩を挙げることができる。

【００１７】前記電解質の前記非水溶媒に対する溶解量は、０．２ｍｏｌ／ｌ～２ｍｏｌ／ｌとすることが望ましい。前記非水電解液を保持するポリマーとしては、例えば、ポリエチレンオキサイド誘導体、ポリプロピレンオキサイド誘導体、前記誘導体を含むポリマー、ビニリデンフロライド（ＶｄＦ）とヘキサフルオロプロピレン（ＨＦＰ）との共重合体等を用いることができる。前記固体電解質層において、このようなポリマーは、分子間が架橋された形態で存在していても良い。また、前記共重合体において、ＶｄＦは共重合体の骨格部で機械的強度の向上に寄与し、ＨＦＰは前記共重合体に非晶質の状態で取り込まれ、非水電解液の保持とリチウムイオンの透過部として機能する。前記ＨＦＰの共重合割合は、前記共重合体の合成方法にも依存するが、通常、最大で２０重量%前後である。|

battery 9 which is formed from theexternally mounted member 8 which was formed it is to be possesses hot seal section 6of frame by 2 film 7a and 7b which have hot melt adhesion resin surface which inposition of periphery side wall vicinity is produced.

[0013] Next, you explain concerning solid polymer electrolyte layer 24, positive electrode collector 25, positive electrode layer 26, the negative electrode collector 27 and negative electrode layer 28 and lengthwise film 1, 2, guide plate 3 and elastic roll 5a, 5b which formthe unit battery 21.

[0014] 1) solid polymer electrolyte layer 24

This polymer electrolyte layer 24 includes polymer which keeps nonaqueous electrolyte solution and this electrolyte solution.

[0015] Aforementioned nonaqueous electrolyte solution is manufactured by melting electrolyte in the nonaqueous solvent. As aforementioned nonaqueous solvent, ethylene carbonate (EC), propylene carbonate (PC), butylene carbonate (BC), the dimethyl carbonate (DMC), diethyl carbonate (DEC), ethyl methyl carbonate (EMC), - butyrolactone (- BL), sulfolane, acetonitrile, the 1,2-dimethoxyethane, 1,3-di methoxy propane, dimethyl ether, tetrahydrofuran (THF) and 2 - methyl tetrahydrofuran etc can belisted. Using and 2 kinds or more mixing, it is good using aforementioned nonaqueous solvent,with alone.

[0016] As aforementioned electrolyte, for example lithium per chlorate ($LiClO_4$), lithium hexafluorophosphate ($LiPF_6$), lithium tetrafluoroborate ($LiBF_4$), the lithium hexafluoroarsenate ($LiAsF_6$), lithium trifluoromethane sulfonate ($LiCF_3SO_3$) and lithium bis-(trifluoromethyl sulfonyl) imide [$LiN(CF_3SO_3)_2$] or other lithium salt can be listed.

[0017] As for dissolved amount for aforementioned nonaqueous solvent of aforementionedelectrolyte, it is desirable to make 0.2 mol/l to 2 mol/l. polymer, vinylidene fluoride (VdF) and copolymer etc of hexafluoropropylene (HFP) which include the for example polyethylene oxide derivative, polypropylene oxide derivative and aforementioned derivative as polymer which keepsthe aforementioned nonaqueous electrolyte solution, can be used. In aforementioned solid electrolyte layer as for this kind of polymer, intermolecular isgood existing with shape which crosslinking is done. In addition, VdF contributes to improvement of mechanical strengthwith skeleton of copolymer in aforementioned copolymer, HFP istaken in by aforementioned copolymer with state of amorphous,functions as permeable part of retention and lithium ion of nonaqueous electrolyte solution. copolymerization ratio of aforementioned

HFP depends on also synthetic method of theaforementioned copolymer, but usually, with maximum it isapproximately a 20 weight %.

【００１８】前記固体電解質層２４は、例えば、以下に説明する方法により作製することができる。

[0018] It can produce aforementioned solid electrolyte layer 24, with method which isexplained below for example .

（１）前記非水電解液を保持するポリマーの溶液を調製し、前記溶液に例えばＤＢＰ（ジブチルフタレート）などの可塑剤を添加し、これを成膜、乾燥した後、これを非水電解液中に浸漬し、前記ポリマーに前記電解液を保持させると共に前記可塑剤と前記電解液を置換することによって前記電解液を含浸させることによりポリマー電解質層を作製する。

(1) As solution of polymer which keeps aforementioned nonaqu eous electrolyte solution ismanufactured, for example DBP (dibutyl phthalate) or other plasticizer is added to aforementioned solution, thefilm formation , after drying this, this is soaked in nonaqueous electrolyte solution, theaforementioned electrolyte solution is kept in aforementioned polymer, polymer electrolyte layer isproduced by impregnating aforementioned electrolyte solution with theaforementioned plasticizer and substituting aforementioned electrolyte solution.

【００１９】（２）前記非水電解液を保持するポリマーの溶液を調製し、前記溶液に例えばＤＢＰ（ジブチルフタレート）などの可塑剤を添加し、これを成膜、乾燥した後、前記可塑剤をエタノール等の溶剤で抽出させる等によって除去し、これに非水電解液を含浸させることによりポリマー電解質層を作製する。

[0019] (2) It manufactures solution of polymer which keeps afo rementionednonaqueous electrolyte solution, adds for example DBP (dibutyl phthalate) or other plasticizer to aforementioned solution, film formation , afterdrying this, aforementioned plasticizer it removes with ethanol or other solvent itextracts with such as, it produces polymer electrolyte layer by impregnating nonaqueous electrolyte solutionin this.

【００２０】（３）前記非水電解液を保持するポリマーの溶液を調製し、これを成膜、乾燥した後、非水電解液を含浸させることによってポリマー電解質層を作製する。前記（１）及び（２）の方法において、可塑剤は、前記固体電解質層の強度等の機械的特性の改善や電解液含浸量を向上させて充放電特性を改善する目的で添加される。前記可塑剤を添加した後、成膜し、前記可塑剤と非水電解液が置換されるようにポリマー電解質層に非水電解液を含浸させることによって、非水電解液を前記ポリマーのみならず前記可塑剤が占めていた空間にも含浸させることができるため、電解液含浸量を増大させることができる。

[0020] (3) Solution of polymer which keeps aforementioned no naqueous electrolyte solution ismanufactured, polymer electrolyte layer is produced film formation , after drying this, byimpregnating nonaqueous electrolyte solution. Regarding to method of aforementioned (1) and (2), plasticizer, improvement and electrolyte solution impregnation amount of strength or other mechanical property of aforementioned solid electrolyte layerimproving, is added with object which improve charge-discharge characteristic. After adding aforementioned plasticizer, film formation it does, in order forthe aforementioned plasticizer and nonaqueous electrolyte solution to be substituted, because the nonaqueous electrolyte solution it can impregnate even in space which aforementionedpolymer furthermore aforementioned plasticizer has occupied byimpregnating nonaqueous electrolyte solution in polymer electrolyte layer, electrolyte solution impregnation amount can be increased.

【００２１】なお、前記（１）～（３）の製造方法において、電解液の含浸（可塑剤添加の場合は可塑剤の除去も含む）は、電解液未含浸の素電池を組み立てた後、或いは図４に示す外装部材８に素電池２１を収納した後でもよい。|

[0021] Furthermore, impregnation (In case of plasticizer additi on also removal of plasticizer includes.) of electrolyte solution after assembling theunit battery of electrolyte solution unimpregnated, or after storing up unit battery 21 in externally mounted member 8 whichis shown in Figure 4 even is good in manufacturing method of aforementioned(1) to (3).

【００２２】２）正極集電体２５

[0022] 2) positive electrode collector 25

この正極集電体２５は、アルミニウムからなるパンチドメタ

As for this positive electrode collector 25, nonaqueous electroly

ルのような非水電解液が流通可能な構造のものを用いることができる。なお、非水電解液が流通可能な構造の正極集電体としては、パンチドメタルの他にアルミニウム製メッシュ、アルミニウム製エキスパンドメタル等を用いることができる。また、前記正極集電体は非水電解液が流通可能な構造のものの他に、アルミニウム箔を用いることができる。|

te solution like punched metal which consists of the aluminum can use those of structure which is circulation possible. Furthermore , aluminum mesh and aluminum expanded metal etc can be used forother than punched metal nonaqueous electrolyte solution as positive electrode collector of structure which iscirculation possible. In addition, aforementioned positive electrode collector nonaqueous electrolyte solution can use aluminum foil forother than those of structure which is circulation possible.

【００２３】３）正極層２６

[0023] 3) positive electrode layer 26

前記正極層２６は、活物質、導電性材料、非水電解液及びこの電解液を保持するポリマーを含む。

Aforementioned positive electrode layer 26 includes polymer which keeps active substance , the electrically conductive material , nonaqueous electrolyte solution and this electrolyte solution.

【００２４】前記活物質としては、種々の酸化物（例えば$LiMn_2O_4$などのリチウムマンガン複合酸化物、二酸化マンガン、例えば$LiNiO_2$などのリチウム含有ニッケル酸化物、例えば$LiCoO_2$などのリチウム含有コバルト酸化物、リチウム含有ニッケルコバルト酸化物、リチウムを含む非晶質五酸化バナジウムなど）や、カルコゲン化合物（例えば、二硫化チタン、二硫化モリブテンなど）等を挙げることができる。中でも、リチウムマンガン複合酸化物、リチウム含有コバルト酸化物、リチウム含有ニッケル酸化物を用いるのが好ましい。

[0024] As aforementioned active substance, various oxide (Includes for example $LiMn_2O_4$ or other lithium manganese composite oxide , manganese dioxide , for example $LiNiO_2$ or other lithium containing nickel oxide , for example $LiCoO_2$ or other lithium containing cobalt oxide , lithium containing nickel cobalt oxide and the lithium such as amorphous vanadium pentoxide which) and, chalcogen compound (Such as for example titanium disulfide and molybdenum disulfide) etc can belisted. It is desirable even among them to use lithium manganese composite oxide , lithium containing cobalt oxide and the lithium containing nickel oxide.

【００２５】前記導電性材料としては、例えば、人造黒鉛、カーボンブラック（例えばアセチレンブラックなど）、ニッケル粉末等を挙げることができる。前記非水電解液及び前記ポリマーは、前述した固体ポリマー電解質層で説明したものと同様なものが用いられる。

[0025] As aforementioned electrically conductive material, for example artificial graphite and carbon black (Such as for example acetylene black), nickel powder etccan be listed. As for aforementioned nonaqueous electrolyte solution and aforementioned polymer, it canuse those which are similar to those which are explained with thesolid polymer electrolyte layer which is mentioned earlier.

【００２６】前記正極２２は、例えば以下に説明する方法によって作製することができる。

[0026] It can produce aforementioned positive electrode 22, with method which isexplained below for example .

（１）前記非水電解液を保持するポリマーの溶液を調製し、前記溶液に例えばＤＢＰ（ジブチルフタレート）などの可塑剤、前記活物質及び前記導電材料を添加した後、これらを混合し、成膜することにより正極層を作製した後、前記正極層と前記集電体とを例えば熱圧着等によって接着する。これを非水電解液中に浸漬することによって前記ポリマーに前記電解液を保持させると共に前記正極層中の可塑剤と前記電解液を置換することによって前記正極層に電解液を含浸させるか、または前記正極層中の前記可塑剤をエタノール等の溶剤で抽出する等によって除去した後、前記正極層に非水電解液を含浸させることにより正極を作製する。

(1) It manufactures solution of polymer which keeps aforementionednonaqueous electrolyte solution, for example DBP (dibutyphthalate) or other plasticizer and aforementioned active substance and after adding theaforementioned electrically conductive material, it mixes these to aforementioned solution, after producing positive electrode layer by film formation doing, aforementioned positive electrode layerand it glues with aforementioned current collector for example thermobonding etc with. As aforementioned electrolyte solution is kept in aforementioned polymer, bysoaking this in nonaqueous electrolyte solution electrolyte solution is impregnated in theaforementioned positive electrode layer with plasticizer in aforementioned positive electrode layer andsubstituting

aforementioned electrolyte solution, Or aforementioned plasticizer in aforementioned positive electrode layer positive electrode isproduced with ethanol or other solvent after removing it extracts with such as, byimpregnating nonaqueous electrolyte solution in aforementioned positive electrode layer.

【００２７】（２）前記非水電解液を保持するポリマーの溶液を調製し、前記溶液に例えばＤＢＰ（ジブチルフタレート）などの可塑剤、前記活物質及び前記導電材料を添加した後、これらを混合し、正極用塗料を調製する。前記正極用塗料を前記集電体に塗工した後、乾燥させる。これを非水電解液中に浸漬することによって前記ポリマーに前記電解液を保持させると共に前記正極層中の可塑剤と前記電解液を置換することによって前記正極層に電解液を含浸させるか、または前記正極層中の前記可塑剤をエタノール等の溶剤で抽出する等によって除去した後、前記正極層に非水電解液を含浸させることにより正極を作製する。

[0027] (2) Solution of polymer which keeps aforementioned no naqueous electrolyte solution ismanufactured, for example DBP (dibutyl phthalate) or other plasticizer and aforementioned active substance and after addingthe aforementioned electrically conductive material, these are mixed to aforementioned solution,the paint for positive electrode is manufactured. After painting paint for aforementioned positive electrode in theaforementioned current collector, it dries. As aforementioned electrolyte solution is kept in aforementioned polymer, bysoaking this in nonaqueous electrolyte solution electrolyte solution is impregnated in theaforementioned positive electrode layer with plasticizer in aforementioned positive electrode layer andsubstituting aforementioned electrolyte solution, Or aforementioned plasticizer in aforementioned positive electrode layer positive electrode isproduced with ethanol or other solvent after removing it extracts with such as, byimpregnating nonaqueous electrolyte solution in aforementioned positive electrode layer.

【００２８】（３）前記非水電解液を保持するポリマーの溶液を調製し、前記溶液に前記活物質及び前記導電材料を添加した後、これらを混合し、成膜することにより正極層を作製した後、前記正極層と前記集電体とを例えば熱圧着等によって接着し、非水電解液を含浸させることによって正極を作製する。

[0028] (3) It manufactures solution of polymer which keeps afo rementionednonaqueous electrolyte solution, aforementioned active substance and after adding aforementionedelectrically conductive material, it mixes these to aforementioned solution, after producing thepositive electrode layer by film formation doing, aforementioned positive electrode layer and it glues withthe aforementioned current collector for example thermobonding etc with it produces positive electrode byimpregnating nonaqueous electrolyte solution.

【００２９】（４）前記非水電解液を保持するポリマーの溶液を調製し、前記溶液に前記活物質及び前記導電材料を添加した後、これらを混合し、正極用塗料を調製する。前記正極用塗料を前記集電体に塗工した後、乾燥させ、非水電解液を含浸させることによって正極を作製する。

[0029] (4) Solution of polymer which keeps aforementioned no naqueous electrolyte solution ismanufactured, aforementioned active substance and after adding theaforementioned electrically conductive material, these are mixed to aforementioned solution, thepaint for positive electrode is manufactured. After painting paint for aforementioned positive electrode in theaforementioned current collector, drying, it produces positive electrode by impregnatingthe nonaqueous electrolyte solution.

【００３０】前記（１）及び（２）の方法において、可塑剤は前記固体電解質層の強度等の機械的特性の改善や電解液含浸量を向上させて充放電特性を改善する目的で添加される。

[0030] Regarding to method of aforementioned (1) and (2), pl asticizerimprovement and electrolyte solution impregnation amount of strength or other mechanical property of aforementioned solid electrolyte layerimproving, is added with objective which improves charge-discharge characteristic.

【００３１】なお、前記（１）～（４）の方法において、電解液の含浸（可塑剤添加の場合は可塑剤の除去も含む）は、電解液未含浸の素電池を組み立てた後、或いは図４に示す外装部材８に素電池２１を収納した後でもよい。

【００３２】４）負極集電体２７

この負極集電体２７は、銅からなるパンチドメタルのような非水電解液が流通可能な構造のものを用いることができる。なお、非水電解液が流通可能な構造の正極集電体としては、パンチドメタルの他に銅製メッシュ、銅製エキスパンドメタル等を用いることができる。また、前記負極集電体は非水電解液が流通可能な構造のものの他に銅箔を用いることができる。

【００３３】５）負極層２８

この負極層２８は、リチウムイオンを吸蔵放出する炭素質材料と、非水電解液と、この電解液を保持するポリマーを含有する。

【００３４】前記リチウムイオンを吸蔵放出する炭素質材料としては、例えば、有機高分子化合物（例えば、フェノール樹脂、ポリアクリロニトリル、セルロース等）を焼成することにより得られるもの、コークスや、ピッチを焼成することにより得られるもの、人造グラファイト、天然グラファイト等に代表される炭素質材料を挙げることができる。中でも、アルゴンガス、窒素ガス等の不活性ガス雰囲気中において、５００℃～３０００℃の温度で、常圧または減圧下にて前記有機高分子化合物を焼成して得られる炭素質材料を用いるのが好ましい。

【００３５】前記非水電解液及び前記ポリマーは、前述した固体ポリマー電解質層で説明したものと同様なものが用いられる。なお、前記負極層２８は人造グラファイト、天然グラファイト、カーボンブラック、アセチレンブラック、ケッチェンブラック、ニッケル粉末、ポリフェニレン誘導体等の導電性材料、オレフィン系ポリマーや炭素繊維等のフィラーを含むことを許容する。

【００３６】前記負極２３は、例えば以下に説明する方法によって作製することができる。

（１）前記非水電解液を保持するポリマーの溶液を調製し、

[0031] Furthermore, regarding to method of aforementioned (1) to (4),the impregnation (In case of plasticizer addition also removal of plasticizer includes.) of electrolyte solution after assembling unit battery of the electrolyte solution unimpregnated, or after storing up unit battery 21 in externally mounted member 8 which is shown inthe Figure 4 even is good.

[0032] 4) negative electrode collector 27

As for this negative electrode collector 27, nonaqueous electrol yte solution like punched metal which consists of the copper can use those of structure which is circulation possible. Furthermore , copper mesh and copper expanded metal etc can be used forother than punched metal nonaqueous electrolyte solution as positive electrode collector of structure which iscirculation possible. In addition, aforementioned negative electrode collector nonaqueous electrolyte solution cai use copper foil forother than those of structure which is circulation possible.

[0033] 5) negative electrode layer 28

This negative electrode layer 28 contains polymer which keeps carbonaceous material and nonaqueous electrolyte solution andthis electrolyte solution which lithium ion intercalation and release are done.

[0034] Are acquired by calcining for example organic polymer compound (Such as for example phenolic resin , polyacrylonitrile and cellulose) intercalation and release is done theaforementioned lithium ion as carbonaceous material which, those which. Those which are acquired by calcining coke and pitch. carbonaceous material which is represented in artificial graphite and natural graphite etc can belisted. With temperature of 500 °C to 3000 °C, calcining aforementioned organic polymer compound underthe ambient pressure or vacuum in in argon gas and nitrogen gas or other inert gas atmosphere, it is desirableeven among them to use carbonaceou material which is acquired.

[0035] As for aforementioned nonaqueous electrolyte solution and aforementioned polymer, it canuse those which are similar to those which are explained with thesolid polymer electrolyte layer which is mentioned earlier. Furthermore , aforementioned negative electrode layer 28 allows fact thatthe artificial graphite , natural graphite , carbon black , acetylene black , Ketjen Black , nickel powder , the polyphenylene derivative or other electrically conductive material , olefin polymer and carbon fiber or other filler are included.

[0036] It can produce aforementioned negative electrode 23, wi th method which isexplained below for example .

(1) It manufactures solution of polymer which keeps aforement

前記溶液に例えばＤＢＰ（ジブチルフタレート）などの可塑剤、前記活物質を添加した後、これらを混合し、成膜することにより負極層を作製した後、前記負極層と前記集電体とを例えば熱圧着等によって接着する。これを非水電解液中に浸漬することによって前記ポリマーに前記電解液を保持させると共に前記負極層中の可塑剤と前記電解液を置換することによって前記負極層に電解液を含浸させるか、または前記負極層中の前記可塑剤をエタノール等の溶剤で抽出する等によって除去した後、非水電解液を含浸させることにより負極を作製する。|

ionednonaqueous electrolyte solution, after adding for example DBP (dibutyl phthalate) or other plasticizer and aforementioned active substance in theaforementioned solution, it mixes these, after producing negative electrode layer by the film formation doing, aforementioned negative electrode layer and it glues with theaforementioned current collector for example thermobonding etc with. As aforementioned electrolyte solution is kept in aforementioned polymer, bysoaking this in nonaqueous electrolyte solution electrolyte solution is impregnated in theaforementioned negative electrode layer with plasticizer in aforementioned negative electrode layer andsubstituting aforementioned electrolyte solution, Or aforementioned plasticizer in aforementioned negative electrode layer negative electrode isproduced with ethanol or other solvent after removing it extracts with such as, byimpregnating nonaqueous electrolyte solution.

【００３７】（２）前記非水電解液を保持するポリマーの溶液を調製し、前記溶液に例えばＤＢＰ（ジブチルフタレート）などの可塑剤、前記活物質を添加した後、これらを混合し、負極用塗料を調製する。前記負極用塗料を前記集電体に塗工した後、乾燥させる。これを非水電解液中に浸漬することによって前記ポリマーに前記電解液を保持させると共に前記負極層中の可塑剤と前記電解液を置換することによって前記負極層に電解液を含浸させるか、または前記負極層中の前記可塑剤をエタノール等の溶剤で抽出する等によって除去した後、非水電解液を含浸させることにより負極を作製する。

[0037] (2) Solution of polymer which keeps aforementioned no naqueous electrolyte solution ismanufactured, after adding for example DBP (dibutyl phthalate) or other plasticizer and aforementioned active substance inthe aforementioned solution, these are mixed, paint for negative electrode ismanufactured. After painting paint for aforementioned negative electrode in theaforementioned current collector, it dries. As aforementioned electrolyte solution is kept in aforementioned polymer, bysoaking this in nonaqueous electrolyte solution electrolyte solution is impregnated in theaforementioned negative electrode layer with plasticizer in aforementioned negative electrode layer andsubstituting aforementioned electrolyte solution, Or aforementioned plasticizer in aforementioned negative electrode layer negative electrode isproduced with ethanol or other solvent after removing it extracts with such as, byimpregnating nonaqueous electrolyte solution.

【００３８】（３）前記非水電解液を保持するポリマーの溶液を調製し、前記溶液に前記活物質を添加した後、これらを混合し、成膜することにより負極層を作製した後、前記負極層と前記集電体とを例えば熱圧着等によって接着する。これに非水電解液を含浸させることによって負極を作製する。

[0038] (3) It manufactures solution of polymer which keeps afo rementionednonaqueous electrolyte solution, after adding aforementioned active substance to aforementionedsolution, it mixes these, after producing negative electrode layer by film formation doing,the aforementioned negative electrode layer and it glues with aforementioned current collectorthe for example thermobonding etc with. negative electrode is produced by impregnating nonaqueous electrolyte solution in this.

【００３９】（４）前記非水電解液を保持するポリマーの溶液を調製し、前記溶液に前記活物質を添加した後、これらを混合し、負極用塗料を調製する。前記負極用塗料を前記集電体に塗工した後、乾燥させ、非水電解液を含浸させることによって負極を作製する。

[0039] (4) Solution of polymer which keeps aforementioned no naqueous electrolyte solution ismanufactured, after adding aforementioned active substance to theaforementioned solution, these are mixed, paint for negative electrode ismanufactured. After painting paint for aforementioned negative electrode in theaforementioned current collector, drying, it produces negative electrode by impregnatingthe nonaqueous electrolyte solution.

【００４０】前記（１）及び（２）の方法において、可塑剤は、前記固体電解質層の強度等の機械的特性の改善や電解液含浸量を向上させて充放電特性を改善する目的で添加される。

[0040] Regarding to method of aforementioned (1) and (2), plasticizer,improvement and electrolyte solution impregnation amount of strength or other mechanical property of aforementioned solid electrolyte layerimproving, is added with objective which improves charge-discharge characteristic.

【００４１】なお、前記（１）～（４）の方法において、電解液の含浸（可塑剤添加の場合は可塑剤の除去も含む）は、電解液未含浸の素電池を組み立てた後、或いは図４に示す外装部材８に素電池２１を収納した後でもよい。

[0041] Furthermore, regarding to method of aforementioned (1) to (4),the impregnation (In case of plasticizer addition also removal of plasticizer includes.) of electrolyte solution after assembling unit battery of the electrolyte solution unimpregnated, or after storing up unit battery 21 in externally mounted member 8 which is shown inthe Figure 4 even is good.

【００４２】また、電解液未含浸の素電池を組み立てた後、或いは図４に示す外装部材８に素電池２１を収納した後に非水電解液を含浸する場合には、前記正極集電体２５および前記負極集電体２７のいずれか一方もしくは両者がパンチドメタルのような非水電解液が流通可能な構造のものを用いることが好ましい。

[0042] In addition, after assembling unit battery of electrolyte solution unimpregnated, when or afterstoring up unit battery 21 in externally mounted member 8 which is shown in Figure 4 thenonaqueous electrolyte solution is impregnated, nonaqueous electrolyte solution any one or both of theaforementioned positive electrode collector 25 and aforementioned negative electrode collector 27 like punched metal usingthose of structure which is circulation possible is desirable.

【００４３】６）長尺フィルム１、２

[0043] 6) lengthwise film 1, 2

この長尺フィルム１、２は、少なくとも対向する面がポリエチレンのような熱融着性樹脂からなる。前記長尺フィルム１、２は、具体的には熱融着性樹脂フィルム単体；熱融着性樹脂フィルムにポリエチレンテレフタレート、ナイロンのような樹脂フィルムまたはＡｌ箔、銅箔のような金属箔をラミネートしたもの；を用いることができる。このようなラミネートフィルムを長尺フィルムとして用いる場合には、前記素電池側に前記熱融着性樹脂フィルムが位置するように配置する。

This lengthwise film 1, 2 consists of hot melt adhesion resin surface which opposes at leastlike polyethylene. Concretely hot melt adhesion resin film unit ; those which laminate metal foil like resin film orthe Al foil and copper foil like polyethylene terephthalate and nylon in hot melt adhesion resin film; youcan use aforementioned lengthwise film 1, 2. When this kind of laminate film it uses, as lengthwise film in order for theaforementioned hot melt adhesion resin film to be located on aforementioned unit battery side, itarranges.

【００４４】７）ガイド板３

[0044] 7) guide plate 3

このガイド板３は、熱融着性樹脂に対して非熱シール性の材料から形成すればよく、特に熱伝導性が良好な銅やアルミニウムのような金属から形成されることが好ましい。

This guide plate 3 vis-a-vis hot melt adhesion resin if from non hot seal characteristicmaterial it should have formed, it is desirable to be formed from themetal like copper and aluminum where especially thermal conductivity issatisfactory.

【００４５】ガイド板は、前述した図１に示す枠状のものに限らず、図５に示すように長尺フィルム２を複数の素電池２１上に重ねた後、この長尺フィルム２上に２つの細長状のガイド板３ａ、３ｂを前記各素電池２１の前記フィルム２の長さ方向に沿う側壁に近接するように配置してもよい。

[0045] As shown in Figure 5 not just those of frame which is shown inthe Figure 1 which is mentioned earlier, after repeating lengthwise film 2 on theunit battery 21 of plural, in order on this lengthwise film 2 2 it is long andnarrow guide plate 3a, 3b, condition proximity to do in side wall whichparallels to longitudinal direction of aforementioned film 2 of theaforementioned each unit battery 21, it is possible to arrange guide plate.

【００４６】８）弾性ロール５ａ、５ｂ

[0046] 8) elastic roll 5a, 5b

これらの弾性ロール５ａ、５ｂは、前記長尺フィルム１、２

As for these elastic roll 5a, 5b, from fact that it is heated in orde

を熱融着するために加熱されることから、前記フィルムの軟化点温度に耐えられるものであるが必要で、例えばシリコーンゴム、フッ素ゴム等を用いることができる。

r hot melt adhesionto do aforementioned lengthwise film 1, 2, i is something which withstands thesoftening point of aforementioned film, but being necessary, you can usethe for example silicone rubber and fluororubber etc.

【００４７】なお、前記弾性ロール５ａ、５ｂ間を通して前記第１、第２の長尺フィルムを互いに熱融着した後、電池形状を安定化させるために熱融着した前記第１、第２の長尺フィルムを加熱されていない別の弾性ロール間に通して冷却することを許容する。|

[0047] Furthermore, aforementioned 1st , second lengthwise fil m mutually hot melt adhesionafter doing, battery shape in order to be stabilized aforementioned1st which hot melt adhesion is done, passing between another elastic roll whichthrough second lengthwise film is not heated through between aforementionedelastic roll 5a, 5b, it allows fact that it cools.

【００４８】以上説明した本発明に係るポリマー電解質二次電池の製造方法は、例えば図１に示すように第１長尺フィルム１の熱融着性樹脂面上に複数の素電池２１を前記フィルム１の長さ方向に所望の間隔をあけて設置する工程と、第２長尺フィルム２の熱融着性樹脂面を複数の前記素電池２１上面を覆うように重ねる工程と、前記第２長尺フィルム２上に厚さが前記素電池２１と同等もしくは僅かに厚い枠状のガイド板３を前記各素電池２１の周囲側壁に近接するようにそれぞれ配置し、前記ガイド板３の底面で前記２枚の長尺フィルム１、２を互いに重ねる工程と、前記複数の素電池２１を前記ガイド板３により間に挟んだ２枚の長尺フィルム１、２を加熱された一対の耐熱性の弾性ゴムロール５ａ、５ｂの間に通過させて前記各素電池２１周辺の前記２枚の長尺フィルム１、２を加熱加圧してそれらの熱融着樹脂面を互いに熱シールする工程とを具備する。

[0048] Above as for manufacturing method of polymer electrol yte secondary battery which relates to this invention whichis explained, as shown in for example Figure 1, step which opening desired intervalto longitudinal direction of aforementioned film 1, installs unit battery 21 of theplural on hot melt adhesion resin surface of 1st lengthwise film 1. step which in order to cover aforementioned unit battery 21 upper surface of theplural, repeats hot melt adhesion resin surface of 2nd lengthwise film 2. In order guide plate 3 of frame where thickness is thick equally tothe aforementioned unit battery 21 or barely on aforementioned 2nd lengthwise film 2 theproximity to do in periphery side wall of aforementioned each unit battery 21, stepwhich it arranges respectively, repeats aforementioned 2 lengthwise film 1, 2mutually with bottom surface of aforementioned guide plate 3. unit battery 21 of aforementioned plural is heated passing 2 lengthwise film 1, 2which put between between with aforementioned guide plate 3 between theelastic rubber roll 5a, 5b of heat resistance of pair which heating and pressurizing doing theaforementioned 2 lengthwise film 1, 2 of aforementioned each unit battery 21 periphery, those hot melt adhesion resin surfacemutually it possesses with step which hot seal is done.

【００４９】このような方法によれば、図４に示すように素電池２１はその周囲に熱シール部６を有する２枚のフィルム７ａ、７ｂからなる外装部材８で覆われ、前記素電池２１の正極、負極側の面に前記フィルム７ａ、７ｂを密着しているため、前記素電池２１と前記外装部材８間に殆ど空隙がない状態で前記素電池２１を収納できる。その結果、前記素電池２１の充放電時に前記素電池２１を構成する正極層２６、負極層２８および固体ポリマー電解質層２４が膨脹・収縮を繰り返しても、前記素電池２１の正極、負極側の面に密着した前記外装部材８の２枚のフィルム７ａ、７ｂにより抑えることができるため、各構成部材の膨脹・収縮によりそれらの中の非水電解液が滲み出するのを抑制できる。特に、前記素電池の正極および負極のいずれか一方もしくは両者にパンチドメタルのような非水電解液が流通可能な構造の集電体を用いた場合、前記素電池２１の各構成部材の膨脹・収縮によりそれらの中の非水電解液がより滲み出し易くなるが、前記外装部材８の２枚のフィルム７ａ、７ｂにより前記滲み出しを効

[0049] According to this kind of method, as shown in Figure 4, as for theunit battery 21 it is covered with externally mounted member 8 which consists of 2 film 7a and the7b which possess hot seal section 6 in periphery because thepositive electrode of aforementioned unit battery 21, aforementioned film 7a, it hasstuck 7b on aspect of negative electrode side, between aforementionedunit battery 21 and aforementioned externally mounted member 8 almost it can store up theaforementioned unit battery 21 with state which does not have gap. As a result, nonaqueous electrolyte solution among those oozing puts out positive electrode layer 26, negative electrode layer 28 and thesolid polymer electrolyte layer 24 which form aforementioned unit battery 21 at time of charge-discharge ofthe aforementioned unit battery 21 expansion * contraction over again, because it ispossible, to hold down positive electroc of aforementioned unit battery 21, the2 film 7a of aforementioned externally mounted member 8 which it sticks

果的に抑制できる。また、前記素電池２１を各構成部材の膨脹・収縮によりそれらの中の非水電解液が滲み出そうとしても、前記素電池２１と前記外装部材８間に殆ど空隙がないため、滲み出し量を低減できる。したがって、充放電の繰り返しによる素電池２１内の非水電解液量の低下を抑制できるため、長期間亘って高い容量を有するポリマー電解質二次電池を得ることができる。

to aspect of the negative electrode side, with 7b with expansior * contraction of each constituting comp[illegible]can control. Especially, when nonaqueous electrolyte solution like punched metal in any one or both ofthe positive electrode and negative electrode of aforementioned unit battery current collector of structure which iscirculation possible is used, nonaqueous electrolyte solution among those it becomes theexuding easier depending upon expansion * contraction of each constituting component of theaforementioned unit battery 21 aforementioned exuding can be controled in theeffective, but 2 film 7a of aforementioned externally mounted member 8, with 7b . In addition, trying, that nonaqueous electrolyte solution among those oozing will put outthe aforementioned unit battery 21 with expansion * contraction of each constituting component,because almost there is not a gap between aforementioned unit battery 21and aforementioned externally mounted member 8, it can decrease exuding quantity. Therefore, because decrease of amount of nonaqueous electrolyte solution inside unit battery 21 due to therepetition of charge-discharge can be controled, long perio extending, it canacquire polymer electrolyte secondary battery which possesses high capacity.

【００５０】また、複数の素電池２１を前記ガイド板３により間に挟んだ２枚の長尺フィルム１、２を加熱された一対の耐熱性の弾性ロール５ａ、５ｂの間に通過させる際、厚さが前記素電池２１と同等もしくは僅かに厚い枠状のガイド板３を前記各素電池２１の周囲側壁に近接するように配置することによって、前記素電池２１に弾性ロール５ａ、５ｂの圧力が加わるのを抑制できる。その結果、前記弾性ロール５ａ、５ｂ間に複数の素電池２１を前記ガイド板３により間に挟んだ２枚の長尺フィルム１、２を通過させる際、前記素電池２１に既に非水電解液が含浸されていても、前記素電池２１からの電解液の滲み出しを抑制することができる。

[0050] In addition, Occasion where it passes between elastic rol 5a, 5b of heat resistance of thepair which 2 lengthwise film 1, 2 which put between unit battery 21 of plural betweenwith aforementioned guide plate 3 is heated, in order guide plate 3 c theframe where thickness is thick equally to aforementioned unit battery 21 orbarely proximity to do in periphery sidewall of aforementioned each unit battery 21,the fact that pressure of elastic roll 5a, 5b joins to aforementioned unit battery 21by arranging, can be controled. As a result, occasion where 2 lengthwise film 1, 2 which put between unit battery 21of plural between between aforementioned elastic roll 5a, 5b with theaforementioned guide plate 3 is passed, nonaqueous electrolyte solution being already impregnated inthe aforementioned unit battery 21, exuding of electrolyte solution from theaforementioned unit battery 21 can be controled.

【００５１】なお、前述した図５に示すように複数の素電池２１上に第２長尺フィルム２を重ねた後、この長尺フィルム２上に２つの細長状のガイド板３ａ、３ｂを前記各素電池２１の前記フィルム２の長さ方向に沿う側壁に近接するように配置した状態で弾性ロール５ａ、５ｂを通過させた場合にも、枠状のガイド板を用いたのとほぼ同様な効果を有するポリマー電解質二次電池を得ることができる。ただし、枠状のガイド板を用いた方が長尺フィルムの長さ方向のみならず、その幅方向に沿う前記素電池の側壁、つまり素電池の周囲側壁にシール性が良好な熱シール部を形成できる。したがって、素電池が収納された外装部材内の空隙量を著しく低減できるため、図５に示す２枚のガイド板３ａ、３ｂを用いる場合より充放電の繰り返しによる素電池２１内の非水電解液量の低下をより効果的に抑制することができる。

[0051]

【００５２】次に、本発明に係る別のポリマー電解質二次電池の製造方法を図６～図８を参照して詳細に説明する。まず、図６に示すように第１長尺フィルム１の熱融着性樹脂面上に前述した構造の複数の矩形状のポリマー電解質素電池２１を前記フィルム１の長さ方向に所望の間隔をあけて設置する。前記各素電池２１を前記第１長尺フィルム１に設置するにあたっては、例えば前記各素電池２１の裏面に接着剤をポイント的に付着させて前記長尺フィルム１に仮固定するすることが好ましい。つづいて、前記長尺フィルム１上に厚さが前記素電池２１と同等もしくは僅かに厚い高分子樹脂からなる枠状のスペーサ１０を前記各素電池２１の周囲側壁に接触するようにそれぞれ設置する。ひきつづき、第２長尺フィルム１の熱融着性樹脂面を前記素電池２１および前記スペーサ１０の上面を覆うように重ねた後、前記複数の素電池２１およびスペーサ１０を間に挟まれた２枚の長尺フィルム１、２を軸４ａ、４ｂを有する耐熱性の弾性ロール５ａ、５ｂの間に通過させる。前記弾性ロール５ａ、５ｂは、長さが前記長尺フィルム１、２の幅よりも十分に長く、かつ所望の加熱源により例えば前記熱融着製樹脂の軟化点以上に加熱されている。図７に示すように前記２枚の長尺フィルム１、２を加熱された前記弾性ロール５ａ、５ｂの間に通過させ、前記素電池２１および前記スペーサ１０が前記弾性ロール５ａ、５ｂの間に移動すると、前記弾性ロール５ａ、５ｂが前記素電池２１および前記スペーサ１０を包み込むように変形し、前記２枚の長尺フィルム１、２が前記弾性ロール５ａ、５ｂからの熱を受け、前記長尺フィルム１、２が前記枠状のスペーサ１０の上下面にそれぞれ熱シールされる。この後、前記弾性ロール５ａ、５ｂ間を通過させた２枚の長尺フィルム１、２を前記スペーサ１０の周縁の沿って切断することにより図８に示すように素電池２１と、この素電池２１を収納し、その周囲側壁に枠状スペーサ１０およびこのスペーサ１０に熱シールされた熱融着性樹脂面を持つ２枚のフィルム７ａ、７ｂとにより形成された外装部材１１とから構成されたポリマー電解質二次電池１２が製造される。

[0052] Next, referring to Figure 6 to Figure 8, you explain manufacturing method of another polymer electrolyte secondary batterywhich relates to this invention in detail. First, as shown in Figure 6, opening desired interval to longitudinal direction o theaforementioned film 1, it installs polymer electrolyte unit battery 21 of rectangle of pluralof construction which is mentioned earlier on hot melt adhesion resin surface of 1st lengthwise film 1. When aforementioned each unit battery 21 is installed in aforementioned1st lengthwise film 1, it is desirab in back surface of for example aforementioned eachunit battery 21 depositing point, temporary affixion to do adhesive in theaforementioned lengthwise film 1 and to do. Continuing, in order to contact periphery side wall of aforementioned eachunit battery 21, it installs spacer 10 of frame which consists of polymer resinwhere thickness is thick equally to aforementioned unit battery 21 or barelyon aforementioned lengthwise film 1 respectively. After in order to cover upper surface of aforementioned unit battery 21 and theaforementioned spacer 10, repeating hot melt adhesion resin surface of continuation and the 2nd lengthwise film 1, unit battery 21 of aforementioned plural and 2 lengthwise film 1, 2 which wasput between spacer 10 between are passed between elastic roll 5a, 5b of theheat resistance which possesses axial 4a 4b. Aforementioned elastic roll 5a, 5b length is long in fully ir comparisonwith width of aforementioned lengthwise film 1, 2, is heated to softening point or higher ofthe for example aforementioned hot melt adhesion make resin at same time by thedesired heat source . As shown in Figure 7, is heated passin aforementioned 2 lengthwise film 1, 2between aforementioned elastic roll 5a, 5b which, When aforementioned unit battery 21 and aforementioned spacer 10 movebetween aforementioned elastic roll 5a, 5b, in order for aforementionedelastic roll 5a, 5b to wrap aforementioned unit battery 21 and aforementioned spacer 10,it becomes deformed, aforementioned 2 lengthwise film 1, 2 receives heat fromthe aforementioned elastic roll 5a, 5b, aforementioned lengthwise film 1, 2 hot seal makesrespectively top and bottom surfaces of spacer 10 of aforementioned frame. As shown in Figure 8 by margin (edge) of aforementioned spacer 10paralleling, cutting off 2 lengthwis film 1, 2 which passes after this and betweenthe aforementionec elastic roll 5a, 5b, it stores up unit battery 21 and this unit battery 21, thepolymer electrolyte secondary battery 12 which is formed from externally mounted member 11 which was formed in periphery side wall bywith 2 film 7a and 7b which have hot melt adhesion resin surface which hot seal isdone in frame spacer 10 and this spacer 10 is produced.

【００５３】前記長尺フィルム１、２は、少なくとも対向する面がポリエチレンのような熱融着性樹脂からなる。前記長尺フィルム１、２は、具体的には熱融着性樹脂フィルム単体：熱融着性樹脂フィルムにポリエチレンテレフタレート、ナイロンのような樹脂フィルムまたはＡｌ箔、銅箔のような金

[0053] Aforementioned lengthwise film 1, 2 consists of hot mel t adhesion resin surface whichopposes at least like polyethylene Concretely hot melt adhesion resin film unit ; those which laminate metal foil like resin film orthe Al foil and copper foil like polyethylene terephthalate and nylon in hot melt

属箔をラミネートしたもの；を用いることができる。このようなラミネートフィルムを長尺フィルムとして用いる場合には、前記素電池側に前記熱融着性樹脂フィルムが位置するように配置する。

adhesion resin film; youcan use aforementioned lengthwise film 1, 2. When this kind of laminate film it uses, as lengthwise film in order for theaforementioned hot melt adhesion resin film to be located on aforementioned unit battery side, itarranges.

【００５４】前記スペーサは、例えばポリエチレンのような熱融着性樹脂またはポリプロピレン、ナイロン等の高分子樹脂等からなる。特に、熱融着性樹脂からなるスペーサは前記弾性ロール５ａ、５ｂによる加熱、加圧時に長尺フィルム１、２に対して良好に熱シールすることが可能になる。

[0054] Aforementioned spacer, consists of hot melt adhesion resin or polypropylene and the nylon or other polymer resin etc like for example polyethylene. Especially, as for spacer which consists of hot melt adhesion resin when heatingand pressurization with aforementioned elastic roll 5a, 5b it becomessatisfactorily hot seal possible vis-a-vis lengthwise film 1, 2 to do.

【００５５】前記スペーサは、前述した図６に示す枠状のものに限らず、図９に示すように長尺フィルム１に複数の素電池２１をその長さ方向に所望の間隔をあけて設置した後、２つの細長状のスペーサ１０ａ、１０ｂを前記各素電池２１の前記フィルム１の長さ方向に沿う側壁にそれぞれ接触するように配置してもよい。このような細長状のスペーサ１０ａ、１０ｂを前記第１長尺フィルム１に設置するにあたっては、前記スペーサ１０ａ、１０ｂの裏面に接着剤をポイント的に付着させて前記長尺フィルム１に仮固定するすることが好ましい。

[0055] Aforementioned spacer as shown in Figure 9 not just those of theframe which is shown in Figure 6 which is mentioned earlier, afteropening desired interval to longitudinal direction, installing unit battery 21 of plural inthe lengthwise film 1, 2 long and narrow in order to contact side wallwhich parallels to longitudinal direction of aforementioned film 1 of theaforementioned each unit battery 21 respectively, may arrange spacer 10a, 10b ofcondition. When this kind of spacer 10a, 10b of condition is installed long andnarrow in aforementioned 1st lengthwise film 1, it is desirable in back surface ofthe aforementioned spacer 10a, 10b depositing point, temporary affixion to do adhesivein aforementioned lengthwise film 1 and to do.

【００５６】前記スペーサには、収納される素電池の正負極の集電体を外部に引き出すための一対の端子を取り付けることを許容する。前記弾性ロール５ａ、５ｂは、前述したのと同様、前記長尺フィルム１、２を熱融着するために加熱されることから、前記フィルムの軟化点温度に耐えられるものであるが必要で、例えばシリコーンゴム、フッ素ゴム等を用いることができる。

[0056] fact that terminal of pair in order to pull outthe current collector of positive/negative electrode of unit battery which is stored up to outside isinstalled is allowed in aforementioned spacer . As for aforementioned elastic roll 5a, 5b, that you mentioned earlier,similarity, from fact that it is heated in order hot melt adhesion to dothe aforementioned lengthwise film 1, 2, it is something which withstands softening pointof aforementioned film, but being necessary, you can use the for example silicone rubber and fluororubber etc.

【００５７】なお、前記弾性ロール５ａ、５ｂ間を通して前記第１、第２の長尺フィルムを前記スペーサ１０に熱融着した後、電池形状を安定化させるために熱融着した前記第１、第２の長尺フィルムを加熱されていない別の弾性ロール間に通して冷却することを許容する。

[0057] Furthermore, aforementioned 1st , second lengthwise film hot melt adhesion afterdoing, battery shape in order to be stabilized aforementioned 1st which hot melt adhesion is done, passing to aforementioned spacer 10 betweenanother elastic roll which through second lengthwise film is not heated through betweenthe aforementioned elastic roll 5a, 5b, it allows fact that it cools.

【００５８】以上説明した本発明に係る別のポリマー電解質二次電池の製造方法は、例えば図６に示すように第１長尺フィルム１の熱融着性樹脂面上に複数の素電池２１を前記フィルム１の長さ方向に所望の間隔をあけて設置する工程と、前記長尺フィルム上１に厚さが前記素電池２１と同等もしくは僅かに厚い高分子樹脂からなる枠状のスペーサを前記各素電池２１の周囲側壁に接触するようにそれぞれ設置する工程と、第２長尺フィルム２の熱融着性樹脂面を複数の前記素電池

[0058] Above as for manufacturing method of another polymer electrolyte secondary battery which relates to this inventionwhich is explained, as shown in for example Figure 6, step which opening thedesired interval to longitudinal direction of aforementioned film 1, installs unit battery 21 ofthe plural on hot melt adhesion resin surface of 1st lengthwise film 1. step which in order to contact periphery side wall of aforementionedeach unit battery 21, installs spacer of frame

２１および前記スペーサ１０の上面を覆うように重ねる工程と、前記複数の素電池２１およびスペーサ１０を間に挟んだ２枚の長尺フィルム１、２を加熱された一対の耐熱性の弾性ロール５ａ、５ｂの間に通過させて前記各素電池周辺の前記２枚の長尺フィルム１、２を前記枠状のスペーサ１０を挟んで加熱加圧して熱シールする工程を具備する。

which consists of thepolymer resin where thickness is thick equally to aforementioned unit battery 21 orbarely in 1 on aforementioned lengthwise film respectively. step which in order to cover upper surface of aforementionedunit battery 21 and aforementioned spacer 10 of plural, repeats hot melt adhesion resin surface ofthe 2nd lengthwise film 2. Is heated passing unit battery 21 of aforementioned plural and 2 lengthwise film 1, 2which put between spacer 10 between between elastic roll 5a, 5b of heat resistance ofthe pair which it possesses step which putting between spacer 10of aforementioned frame, heating and pressurizing doing, hot seal does theaforementioned 2 lengthwise film 1, 2 of aforementioned each unit battery periphery.

【００５９】このような方法によれば、図８に示すように素電池２１はその周囲側壁に枠状スペーサ１０およびこのスペーサ１０に熱融着性樹脂面が熱シールされた２枚のフィルム７ａ、７ｂとにより形成されたに外装部材１１で覆われ、前記素電池２１の正極、負極側の面に前記フィルム７ａ、７ｂが密着されているため、前記素電池２１と前記外装部材１１間に殆ど空隙がない状態で前記素電池２１を収納できる。その結果、前記素電池２１の充放電時に前記素電池２１を構成する正極層、負極層および固体ポリマー電解質層が膨脹・収縮を繰り返しても、前記素電池２１の正極、負極側の面に密着した前記外装部材１１の２枚のフィルム７ａ、７ｂにより抑えることができるため、各構成部材の膨脹・収縮によりそれらの中の非水電解液が滲み出するのを抑制できる。特に、前記素電池の正極および負極のいずれか一方もしくは両者にパンチドメタルのような非水電解液が流通可能な構造の集電体を用いた場合、前記素電池２１の各構成部材の膨脹・収縮によりそれらの中の非水電解液がより滲み出し易くなるが、前記外装部材１１の２枚のフィルム７ａ、７ｂにより前記滲み出しを効果的に抑制できる。また、前記素電池２１を各構成部材の膨脹・収縮によりそれらの中の非水電解液が滲み出そうとしても、前記素電池２１と前記外装部材１１間に殆ど空隙がないため、滲み出し量を低減できる。したがって、充放電の繰り返しにる素電池２１内の非水電解液量の低下を抑制できるため、長期間亘って高い容量を有するポリマー電解質二次電池を得ることができる。

[0059] In this kind of method we depend, As shown in Figure 8 unit battery 21 in periphery side wall hot melt adhesion resin surface hot seal wasdone in frame spacer 10 and this spacer 10 : film 7a, It was formed by with 7b with externally mounted member 11 to be covered, becausethe positive electrode of aforementioned unit battery 21, aforementioned film 7a, the7b has stuck on aspect of negative electrode side, between aforementionedunit battery 21 and aforementioned externally mounted member 11 almost aforementioned unit battery 21 canbe stored up with state which does not have gap. As a result nonaqueous electrolyte solution among those oozing puts out positive electrode layer , negative electrode layer and thesolid polymer electrolyte layer which form aforementioned unit battery 21 at time of charge-discharge ofthe aforementioned unit battery 21 expansion * contraction over again, because it ispossible, to hold down positive electrode of aforementioned unit battery 21, the2 film 7a of aforementioned externally mounted member 11 which it sticks to aspect of the negative electrode side, with 7b with expansion * contraction of each constituting compon る youcan control. Especially, when nonaqueous electrolyte solution like punched metal in any one or both ofthe positive electrode and negative electrode of aforementioned unit battery current collector of structure which iscirculation possible is used, nonaqueous electrolyte solution among those it becomes theexuding easier depending upon expansion * contraction of each constituting component of theaforementioned unit battery 21 aforementioned exuding can be controled in theeffective, but 2 film 7a of aforementioned externally mounted member 11, with 7b . In addition, trying, that nonaqueous electrolyte solution among those oozing will put outthe aforementioned unit battery 21 with expansion * contraction of each constituting component,because almost there is not a gap between aforementioned unit battery 21and aforementioned externally mounted member 11, it can decrease exuding quantity. Therefore, be る decrease of amount of nonaqueous electrolyte solution inside unit battery 21 can becontroled in repetition of charge-discharge, long period extending, it canacquire polymer electrolyte secondary battery which possesses high capacity.

【００６０】また、複数の素電池２１および前記枠状のスペーサ１０を間に挟んだ２枚の長尺フィルム１、２を加熱された一対の耐熱性の弾性ロール５ａ、５ｂの間に通過させる際、厚さが前記素電池２１と同等もしくは僅かに厚い枠状のスペーサ１０を前記各素電池２１の周囲側壁に接触して配置されているため、前記素電池２１に弾性ロール５ａ、５ｂの圧力が加わるのを抑制できる。その結果、前記弾性ロール５ａ、５ｂ間に複数の素電池２１および前記スペーサ１０を間に挟んだ２枚の長尺フィルム１、２を通過させる際、前記素電池２１に既に非水電解液が含浸されていても、前記素電池２１からの電解液の滲み出しを抑制することができる。

【００６１】さらに、外装部材１１は周囲に枠状のスペーサ１０を有するため、外部からの衝撃を前記スペーサで緩和できるため、信頼性の高いポリマー電解質二次電池を得ることができる。

【００６２】なお、前述した図９に示すように第１長尺フィルム１に複数の素電池２１をその長さ方向に所望の間隔をあけて設置し、２つの細長状のスペーサ１０ａ、１０ｂを前記各素電池２１の前記フィルム１の長さ方向に沿う側壁にそれぞれ接触するように設置した後、前記素電池２１およびスペーサ１０ａ、１０ｂ上に第２長尺フィルム２を重ねた状態で弾性ロール５ａ、５ｂを通過させた場合にも、枠状のスペーサを用いたのとほぼ同様な効果を有するポリマー電解質二次電池を得ることができる。ただし、枠状のスペーサを用いた方が長尺フィルムの長さ方向のみならず、その幅方向に沿う前記素電池の側壁、つまり素電池の周囲側壁に接触するスペーサを有する外装部材を形成できることから、図９に示す２枚のスペーサ１０ａ、１０ｂを用いる場合より素電池と外装部材の間の空隙を極めて小さくでき、充放電の繰り返しによる素電池２１内の非水電解液量の低下をより効果的に抑制することができる。

【００６３】

【実施例】以下、本発明の実施例を前述した図面を参照して詳細に説明する。

（実施例１）

[0060] In addition, Occasion where it passes between elastic r
5a, 5b of heat resistance of thepair which unit battery 21 of
plural and 2 lengthwise film 1, 2 which put between thespacer
10 of aforementioned frame between is heated, because spacer
10of frame where thickness is thick equally to
aforementionedunit battery 21 or barely contacting periphery
sidewall of aforementioned each unit battery 21,it is arranged,
you can control fact that pressure of elastic roll 5a, 5bjoins to
aforementioned unit battery 21. As a result, occasion where
unit battery 21 of plural and 2 lengthwise film 1, 2which put
between aforementioned spacer 10 between are passed
betweenthe aforementioned elastic roll 5a, 5b, nonaqueous
electrolyte solution being already impregnated in
theaforementioned unit battery 21, exuding of electrolyte
solution from aforementionedunit battery 21 can be controled.

[0061] Furthermore, externally mounted member 11 in order t
o possess spacer 10 of frame inthe periphery, because impact
from outside can be eased with theaforementioned spacer, can
acquire polymer electrolyte secondary battery where reliability
is high.

[0062] Furthermore, As shown in Figure 9 which you mention e
arlier, opening desired interval tothe longitudinal direction, it
installs unit battery 21 of plural in 1st lengthwise film 1, When
2 long and narrow in order to contact side wall whichparallels t
longitudinal direction of aforementioned film 1 of
theaforementioned each unit battery 21 respectively with
aforementioned unit battery 21and state which repeats 2nd
lengthwise film 2 on spacer 10a, 10b after, installingthe spacer
10a, 10b of condition, elastic roll 5a, 5b is passed even, spacer
of the frame was used with can acquire polymer electrolyte
secondary battery which almost possessessimilar effect.
However, Method which uses spacer of frame longitudinal
direction of lengthwise filmfurthermore, When side wall of
aforementioned unit battery which parallels to thelateral
direction, in other words from fact that externally mounted
member which possesses thespacer which contacts periphery
side wall of unit battery can be formed, 2 spacer 10a, 10bwhich
is shown in Figure 9 is used compared to gap between theunit
battery and externally mounted member quite it can make small
it can control from thedecrease of amount of nonaqueous
electrolyte solution inside unit battery 21 due to repetition of
the charge-discharge in effective.

[0063]

[Working Example(s)] Below, referring to drawing which menti
ons earlier Working Example of the this invention, you explain
in detail.

(Working Example 1)

<正極の作製>アセトン２０ｇにビニリデンフロライドーヘキサフルオロプロピレン（ＶｄＦ－ＨＦＰ）の共重合体（エルファトケム社製商品名：ＫＹＮＡＲ２７５０、共重合比［ＶｄＦ：ＨＦＰ］が８５：１５）粉末２．８ｇを溶解した後、このアセトン溶液にジブチルフタレート（ＤＢＰ）４．３ｇを添加し、活物質として組成式が$LiCoO_2$で表されるリチウム含有コバルト酸化物を１０．５ｇと、導電性材料としてのアセチレンブラック１．１３ｇを添加し、混合、分散させて正極用ペーストを調製した。前記正極用ペーストをアルミニウム製パンチドメタル（穴径：１．５ｍｍ、開孔率４０％）からなる正極集電体にナイフコータを用いて２．５ｍＡｈ／cm^2となるよう塗布速度１ｍ／ｍｉｎで塗工し、乾燥空気により塗膜を乾燥することによって正極を作製した。

<Production of positive electrode> After melting copolymer (Elf Atochem supplied tradename ; KYNAR2750 and copolymerization ratio [VdF : HFP] 85:15) powder 2.8g of vinylidene fluoride - hexafluoropropylene (VdF - HFP) in acetone 20g,it added dibutyl phthalate (DBP)4.3g to this acetone solution, composition formula being $LiCoO_2$ as the active substance, it added acetylene black 1.13g, lithium containing cobalt oxide which is displayed as 10.5gand electrically conductive material it mixed, dispersed and manufactured paste for thepositive electrode. paste for aforementioned positive electrode in order to become 2.5 mAh/cm2 inthe positive electrode collector which consists of aluminum punched metal (hole diameter ; 1.5 mm and open pore ratio 40 %) making use of knife coater waspainted with application rate 1 m/min, positive electrode was produced by drying coatingwith dry air .

<負極の作製>前記正極層に用いられたのと同様なビニリデンフロライドーヘキサフルオロプロピレンの共重合体２．０ｇをアセトン１２ｇに溶解させた後、このアセトン溶液にジブチルフタレート（ＤＢＰ）３．１２ｇを添加し、活物質としてメソフェーズピッチ系炭素繊維（株式会社ペトカ社製）７．３７ｇを添加し、これらを混合することにより負極用ペーストを調製した。前記負極用ペーストを銅製パンチドメタル（穴径：１．５ｍｍ、開孔率４０％）からなる負極集電体に２．５ｍＡｈ／cm^2となるようナイフコータを用いて前述したのと同様な条件で塗工し、乾燥空気により塗膜を乾燥することによって負極を作製した。

<Production of negative electrode> It was used for aforementio ned positive electrode layer, that copolymer 2.0g of thesimilar vinylidene fluoride - hexafluoropropylene after melting, dibutyl phthalate (DBP)3.12g was added to thisacetone solution in acetone 12g, mesophase pitch type carbon fiber (Petoca, Ltd. supplied)7.37g was added as active substance, paste forthe negative electrode was manufactured by mixing these. paste fo aforementioned negative electrode way it becomes 2.5 mAh/cm2 in thenegative electrode collector which consists of copper punched metal (hole diameter ; 1.5 mm and open pore ratio 40 %) that you mentioned earlier makinguse of knife coater was painted with similar condition, negative electrode wasproduced by drying coating with dry air .

<固体ポリマー電解質層の作製>前記正極層に用いられたのと同様なビニリデンフロライドーヘキサフルオロプロピレンとの共重合体２．０ｇをアセトン１０ｇに溶解させた後、このアセトン溶液にジブチルフタレート（ＤＢＰ）２．０ｇを添加し、混合することによって電解質層用ペーストを調製した。このペーストを平滑なガラス板上に乾燥後の膜厚が７０μｍになるように塗布した後、乾燥し、前記ガラス板から剥離し、固体ポリマー電解質層を作製した。

<Production of solid polymer electrolyte layer> It was used for aforementioned positive electrode layer, that copolymer 2.0g of thesimilar vinylidene fluoride - hexafluoropropylene after melting, dibutyl phthalate (DBP)2.0g was added to thisacetone solution in acetone 10g, paste for electrolyte layer was manufactured bymixing. After in order on smooth glass sheet for film thickness after drying to become the70 m, applying this paste, it dried, peeled off from theaforementioned glass sheet, produced solid polymer electrolyte layer.

<非水電解液の調製>エチレンカーボネート（ＥＣ）とジメチルカーボネート（ＤＭＣ）が体積比で１：１の割合で混合された非水溶媒に電解質としての$LiPF_6$をその濃度が１ｍｏｌ／ｌになるように溶解させて非水電解液を調製した。

<Manufacturing nonaqueous electrolyte solution> Ethylene carl onate (EC) and dimethyl carbonate (DMC) being volume ratio, in order for concentration tobecome 1 mol/l, melting LiPF6 as electrolyte in nonaqueous solvent which ismixed at ratio of 1:1, it manufactured nonaqueous electrolyte solution.

<素電池の作製>得られた正極および負極を２ｃｍ×２ｃｍの大きさに切り出し、前記固体ポリマー電解質層を２．２５ｃｍ×２．２５ｃｍの大きさに切り出した。前記正極と前記負極の間に前記ポリマー電解質層を介在させ、これらを１３０℃に加熱した剛性ロールにて加熱圧着して電解液未含浸の素電池を得た。つづいて、前記電解液未含浸の素電池を前記

<Production of unit battery> Positive electrode and negative ele ctrode which it acquires was cut in size of 2 cm X 2 cm, theaforementioned solid polymer electrolyte layer was cut in size of 2.25 cm X 2.25 cm. Aforementioned polymer electrolyte layer lying between between aforementionedpositive electrode and aforementioned negative electrode,

非水電解液中に２時間浸漬して前記素電池を構成する正極層、負極層およびポリマー電解質層中のＤＢＰと前記電解液を置換して前記電解液を含浸することにより図２に示す構造の素電池を作製した。|

thermobonding doing with stiffness roll which heats these to 130 °C, it acquired unit battery of electrolyte solution unimpregnated. Continuing, 2 hours soaking unit battery of aforementioned electrolyte solution unimpregnated inthe aforementioned nonaqueous electrolyte solution, substituting DBP and theaforementioned electrolyte solution in positive electrode layer , negative electrode layer and polymer electrolyte layer which formthe aforementioned unit battery, it produced unit battery of structure which itshows in Figure 2 by impregnating aforementioned electrolyte solution.

【００６４】次いで、前述した図１に示すように厚さ５０μｍのポリエチレン（ＰＥ）フィルムと厚さ５０μｍのポリエチレンテレフタレート（ＰＥＴ）フィルムとをラミネートした第１長尺フィルム１のＰＥフィルム上に前述した方法で作製した複数の素電池２１を前記フィルム１の長さ方向に所望の間隔をあけてそれぞれ設置すると共に前記素電池裏面にポイント的に付けた接着剤により仮固定した。厚さ５０μｍのＰＥフィルムと厚さ５０μｍのＰＥＴフィルムとをラミネートした第２長尺フィルム２のＰＥフィルム面を複数の前記素電池２１上面を覆うように重ねた後、前記第２長尺フィルム２上に厚さが前記素電池２１より僅かに厚い銅からなる枠状のガイド板３を前記素電池２１の周囲側壁に近接するように配置し、前記ガイド板３の底面で前記２枚の長尺フィルム１、２を互いに重ねた。つづいて、前記複数の素電池２１が前記ガイド板３により間に挟まれた２枚の長尺フィルム１、２を軸４ａ、４ｂを有するシリコーンゴムからなる弾性ロール５ａ、５ｂの間に通過させた。前記弾性ロール５ａ、５ｂは、長さが前記長尺フィルム１、２の幅よりも十分に長く、かつ所望の加熱源により１１０℃に加熱されている。図３に示すように前記２枚の長尺フィルム１、２を加熱された前記弾性ロール５ａ、５ｂの間に通過させ、前記素電池２１および前記ガイド板３が前記弾性ロール５ａ、５ｂの間に移動させることにより、前記弾性ロール５ａ、５ｂが前記素電池２１および前記ガイド板３を包み込むように変形し、前記ガイド板３の底面で互いに重ねられた前記２枚の長尺フィルム１、２部分が互いに熱シールされた。ひきつづき、前記弾性ロール５ａ、５ｂ間を通過させた２枚の長尺フィルム１、２から前記ガイド板３を外し、前記２枚の長尺フィルム１、２を前記ガイド板３周縁が位置された箇所に沿って切断することにより図４に示すように素電池２１とこの素電池２１を収納し、その周囲側壁付近に位置した枠状の熱シール部６を有する２枚のＰＥ／ＰＥＴのラミネートフィルム７ａ、７ｂにより形成された外装部材８とから構成されたポリマー電解質二次電池９を製造した。|

[0064] Next, as shown in Figure 1 which is mentioned earlier, a opening thedesired interval to longitudinal direction of aforementioned film 1, it installs unit battery 21of plural which is produced with method which is mentionedearlier on PE film of 1st lengthwise film 1 which laminates with polyethylene (PE film ofthe thickness 50 m and polyethylene terephthalate (PET) film of thickness 50 m respectively temporary affixion it didwith adhesive which you attach to aforementioned unit battery back surface point. In order PE film surface of 2nd lengthwise film 2 which laminates with PE film of the thickness 50 m and PET film of thickness 50 m to cover aforementioned unit battery 21 upper surface ofthe plural, after repeating, in order proximity to do guide plate 3 of theframe which consists of copper where thickness is barely thickerthan aforementioned unit battery 21 on aforementioned 2nd lengthwise film 2 in periphery side wallof aforementioned unit battery 21, it arranged, repeated aforementioned2 lengthwise film 1, 2 mutually with bottom surface of aforementioned guide plate 3. Continuing, unit battery 21 of aforementioned plural it passed 2 lengthwise film 1, 2which was put between between by aforementioned guide plate 3 between theelastic roll 5a, 5b which consists of silicone rubber which possesses axial 4a, 4b. Aforementioned elastic ro 5a, 5b length is long in fully in comparisonwith width of aforementioned lengthwise film 1, 2, is heated to 110 °C atthe same time by desired heat source . As shown in Figure 3, is heated passing aforementioned 2 lengthwise film 1, 2between aforementioned elastic roll 5a, 5b which, in order for theaforementioned elastic roll 5a, 5b to wrap aforementioned unit battery 21 and theaforementioned guide plate 3 due to fact that aforementioned unit battery 21 andthe aforementioned guide plate 3 move between aforementioned elastic roll 5a, 5b, itbecame deformed, aforementioned 2 lengthwise film 1, 2 portion which is repeated mutuallywith bottom surface of aforementioned guide plate 3 was done hot sealmutually. Continuation, From 2 lengthwise film 1, 2 which passes betwee aforementioned elastic roll 5a, 5b theaforementioned guide plate 3 to remove, As shown in Figure 4 by cutting off aforementioned 2 lengthwise film 1, 2alongside site where aforementioned guide plate 3 peripheral edge is located, theunit battery 21 and this unit battery 21 were stored up, polymer electrolyte secondary battery 9 which is formed fromthe

【００６５】（実施例２）まず、図６に示すように厚さ５０μｍのＰＥフィルムと厚さ５０μｍのＰＥＴフィルムとをラミネートした第１長尺フィルム１のＰＥフィルム上に実施例１と同様な素電池２１を前記フィルム１の長さ方向に所望の間隔をあけてそれぞれ設置すると共に前記素電池裏面にポイント的に付けた接着剤により仮固定した。つづいて、前記長尺フィルム１上に厚さが前記素電池２１と同等のポリエチレンからなる枠状のスペーサ１０を前記素電池２１の周囲側壁に接触するようにそれぞれ設置した。厚さ５０μｍのＰＥフィルムと厚さ５０μｍのＰＥＴフィルムとをラミネートした第２長尺フィルム２のＰＥ面を前記素電池２１および前記スペーサ１０の上面を覆うように重ねた後、前記複数の素電池２１およびスペーサ１０を間に挟まれた２枚の長尺フィルム１、２を軸４ａ、４ｂを有するシリコーゴムからなる弾性ロール５ａ、５ｂの間に通過させた。前記弾性ロール５ａ、５ｂは、長さが前記長尺フィルム１、２の幅よりも十分に長く、かつ所望の加熱源により１１０℃に加熱されている。図７に示すように前記２枚の長尺フィルム１、２を加熱された前記弾性ロール５ａ、５ｂの間に通過させ、前記素電池２１および前記スペーサ１０を前記弾性ロール５ａ、５ｂの間に移動させることにより、前記弾性ロール５ａ、５ｂが前記素電池２１および前記スペーサ１０３を包み込むように変形し、前記長尺フィルム１、２のＰＥフィルムが前記枠状のスペーサ１０の上下面にそれぞれ熱シールされた。ひきつづき、前記弾性ロール５ａ、５ｂ間を通過させた２枚の長尺フィルム１、２を前記スペーサ１０の周縁の沿って切断することにより図８に示すように素電池２１と、この素電池２１を収納し、その周囲側壁に枠状スペーサ１０およびこのスペーサ１０に熱シールされた２枚のＰＥ／ＰＥＴのラミネートフィルム７ａ、７ｂとにより形成された外装部材１１とから構成されたポリマー電解質二次電池１２を製造した。

【００６６】（比較例）内容積が実施例１と同様な素電池に近似し、一辺が開口した厚さ５０μｍのＰＥフィルムと厚さ５０μｍのＰＥＴフィルムとをラミネートした構造の未密封袋を作製し、この未密封袋の開口部から前記素電池を挿入し、前記開口部を熱シールすることにより外装部材である密封袋内に素電池が収納されたポリマー電解質二次電池を製造した。

externally mounted member 8 which was formed it is to be possesses hot seal section6 of frame laminate film 7a of 2 PE/ PET which, by 7b inposition of periphery side wall vicinity was produced.

[0065] (Working Example 2) First, as shown in Figure 6, as opening desired interval to longitudinal direction of theaforementioned film 1, it installs unit battery 21 which is similar to theWorking Example 1 in PE film of thickness 50 m and on PE film of 1st lengthwise film 1 whichlaminates PET film of thickness 50 m respectively temporary affixion it did withthe adhesive which you attach to aforementioned unit battery back surface point. Continuing, in order to contact periphery side wall of aforementioned unit battery 21,it installed spacer 10 of frame which consists of polyethylene wherethe thickness is equal to aforementioned unit battery 21 on aforementionedlengthwise film 1 respectively. After in order to cover upper surface of aforementioned unit battery 21 and theaforementioned spacer 10, repeating P E face of 2nd lengthwise film 2 which laminateswith PE film of thickness 50 m and PET film of thickness 50 m, unit battery 21 ofthe aforementioned plural and 2 lengthwise film 1, 2 which was put between spacer 10between were passed between elastic roll 5a, 5b which consists of silico -rubber which possesses axial 4a, 4b Aforementioned elastic roll 5a, 5b length is long in fully in comparisonwith width of aforementioned lengthwise film 1, 2, is heated to 110 °C atthe same time by desired heat source . As shown in Figure 7, is heated passing aforementioned 2 lengthwise film 1, 2between aforementioned elastic roll 5a, 5b which, in order for theaforementioned elastic roll 5a, 5b to wrap aforementioned unit battery 21 and theaforementioned spacer 103 with aforementioned unit battery 21 and moving theaforementioned spacer 10 between aforementioned elastic roll 5a, 5b, it becamedeformed, PE film of aforementioned lengthwise film 1, 2 hot seal maderespectively top and bottom surfaces of spacer 10 of aforementioned frame. As shown in Figure 8 by surrounding edge of aforementioned spacer 10paralleling, cutting off 2 lengthwise film 1, 2 which passes during continuingand aforementioned elastic roll 5a, 5b, it stored up unit battery 21 and this unit battery 21,it produced polymer electrolyte secondary battery 12 which is formed from externally mounted member 11 which was formedin periphery side wall laminate film 7a of 2 PE/ PET which hot seal is done, by withthe 7b in frame spacer 10 and this spacer 10.

[0066] (Comparative Example) It closely resembled to unit battery where internal volume is similar to theWorking Example 1, it produced not yet sealed bag of structure which laminates withthe PE film of thickness 50 m which one edge opens and PET film of thethickness 50 m, inserted aforementioned unit battery from opening part of this notyet sealed bag, it produced polymer electrolyte secondary battery where unit

【００６７】得られた実施例１、２および比較例の二次電池について、収納された素電池の正極集電体および負極集電体をリードを用いて外部に引き出し、充電電流２ｍＡ、４．２Ｖ、１０時間の定電流定電圧充電を行った後、２．７Ｖまで２ｍＡの電流で放電する充放電を繰り返し行った。１００回の充放電を繰り返した後の初期容量に対する放電容量維持率を測定した。その結果を下記表１に示す。

【００６８】

表１

	放電容量維持率（%）
実施例１	８５
実施例２	７０
比較例	１７

前記表１から明らかなように本実施例１、２により得られた二次電池は、比較例の二次電池に比べて放電容量維持率が極めて高いことがわかる。これは、実施例１、２の二次電池の外装フィルム、外装部材を構成する２枚のＰＥ／ＰＥＴのラミネートフィルムが素電池の正極、負極側の面に密着していること、素電池が収納された外装部材内の殆ど空隙が生じていないことに起因する。

【００６９】

【発明の効果】以上詳述したように本発明によれば、外装部材とこの中に収納された素電池との間の空隙量を著しく低減して充放電時における前記素電池の膨脹・収縮に起因する非水電解液の滲み出しを抑制し、ひいては長期間亘って高い容量を有するポリマー電解質二次電池の製造方法を提供することができる。

【図面の簡単な説明】

【図１】本発明に係るポリマー電解質二次電池の製造を説明するための斜視図。

battery is stored up insidethe sealed bag which is a externally mounted member by hot seal doing aforementionedopening part.

[0067] Concerning secondary battery of Working Example 1, 2 and Comparative Example which it acquires, the positive electrode collector and negative electrode collector of unit battery which is stored up werepulled out to outside making use of lead, after doing constant current constant voltage charging of charging current 2 mA , 4.2V and 10 hours, charge-discharge which discharges withthe current of 2 mA repeatedly was done to 2.7V. After repeating charge-discharge of 100 tim discharge capacity maintenance ratio for initial capacitywas measured. Result is shown in below-mentioned Table 1.

[0068]

As been clear from aforementioned Table 1, as for secondary ba ttery which isacquired with this working example 1, 2 , it understands that discharge capacity maintenance ratio quite is highin comparison with secondary battery of Comparative Example. external mounting film of secondary battery of Working Example 1, 2, laminate film of 2 PE/ PET whichforms externally mounted member positive electrode of unit battery, has stuck this, to aspectof negative electrode side, it originates in inside externally mounted member where unit battery isstored up almost gap not occurring.

[0069]

[Effects of the Invention] As above detailed, according to this i nvention, decreasing void amount withthe externally mounted member and unit battery which among these is stored up considerably,you control exuding of nonaqueous electrolyte solution which originates in expansion *contraction of aforementioned unit battery at time of charge-discharge, the consequently long period extend and you can offer manufacturing method of polymer electrolyte secondary battery which possessesthe high capacity.

[Brief Explanation of the Drawing(s)]

[Figure 1] Oblique view in order to explain production of polyr er electrolyte secondary battery whichrelates to this invention.

【図２】本発明に用いられる素電池の部分切欠斜視図。

【図３】図１の弾性ロール付近の部分拡大断面図。

【図４】図１に示す方法により製造されたポリマー電解質二次電池の部分切欠斜視図。

【図５】本発明に係るポリマー電解質二次電池の他の製造形態を説明するための斜視図。

【図６】本発明に係る別のポリマー電解質二次電池の製造を説明するための斜視図。

【図７】図６の弾性ロール付近の部分拡大断面図。

【図８】図６に示す方法により製造されたポリマー電解質二次電池の部分切欠斜視図。

【図９】本発明に係る別のポリマー電解質二次電池における他の製造形態を説明するための斜視図。

【符号の説明】

１…第１長尺フィルム、

２…第２長尺フィルム、

３、３ａ、３ｂ…ガイド板、

５ａ、５ｂ…弾性ロール、

６…熱シール部、

７ａ、７ｂ…熱融着性樹脂面を持つフィルム

８、１１…外装部材、

９、１２…ポリマー電解質二次電池、

１０、１０ａ、１０ｂ…スペーサ、

２１…素電池、

２２…正極、

[Figure 2] Partially cut-out oblique diagram of unit battery which is used for this invention.

[Figure 3] Portion enlarged cross section diagram of elastic roll vicinity of Figure 1.

[Figure 4] Partially cut-out oblique diagram of polymer electrolyte secondary battery which is produced by method which is shownin Figure 1.

[Figure 5] Oblique view in order to explain other production form of polymer electrolyte secondary battery whichrelates to this invention.

[Figure 6] Oblique view in order to explain production of another polymer electrolyte secondary battery whichrelates to this invention.

[Figure 7] Portion enlarged cross section diagram of elastic roll vicinity of Figure 6.

[Figure 8] Partially cut-out oblique diagram of polymer electrolyte secondary battery which is produced by method which is shownin Figure 6.

[Figure 9] Oblique view in order to explain other production form in another polymer electrolyte secondary battery whichrelates to this invention.

[Explanation of Reference Signs in Drawings]

1... 1st lengthwise film ,

2... 2nd lengthwise film ,

3 and 3a, 3b ... guide plate ,

5a, 5b ... elastic roll ,

6... hot seal section,

It has 7a and 7b... hot melt adhesion resin surface film

8 and 11... externally mounted member ,

9, 12... polymer electrolyte secondary battery ,

10, 10a and 10b... spacer ,

21... unit battery ,

22... positive electrode ,

２３…負極、

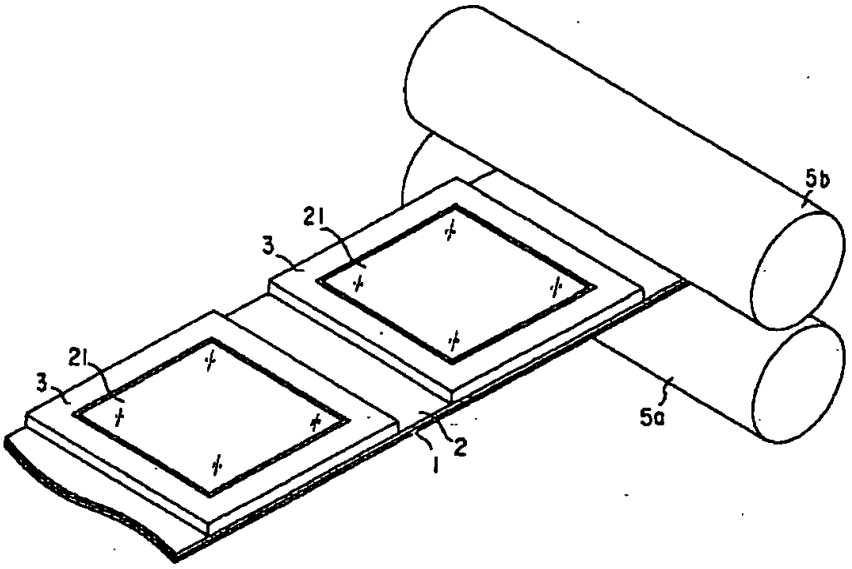
２４…固体ポリマー電解質層。

23... negative electrode ,

24... solid polymer electrolyte layer .

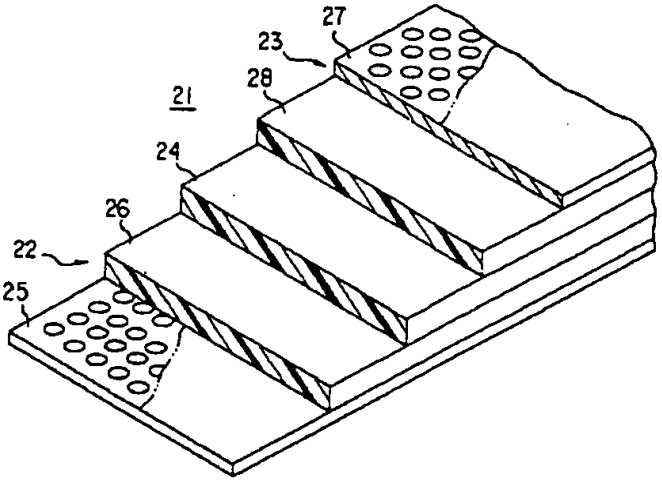
【図１】

[Figure 1]

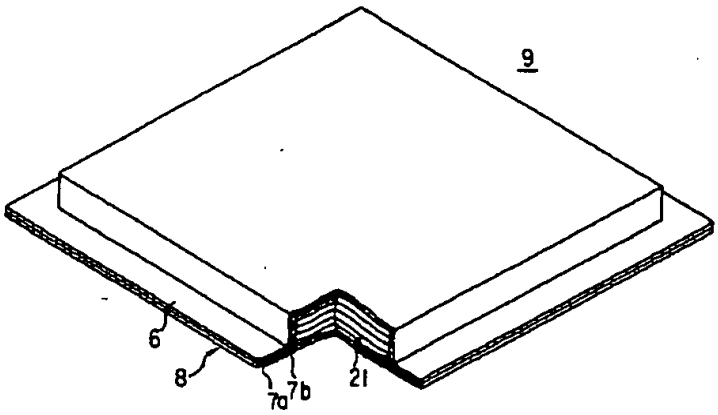


【図２】

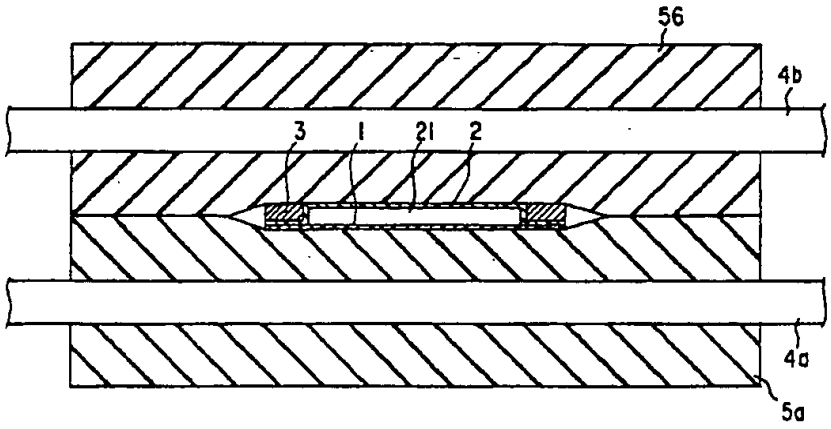
[Figure 2]



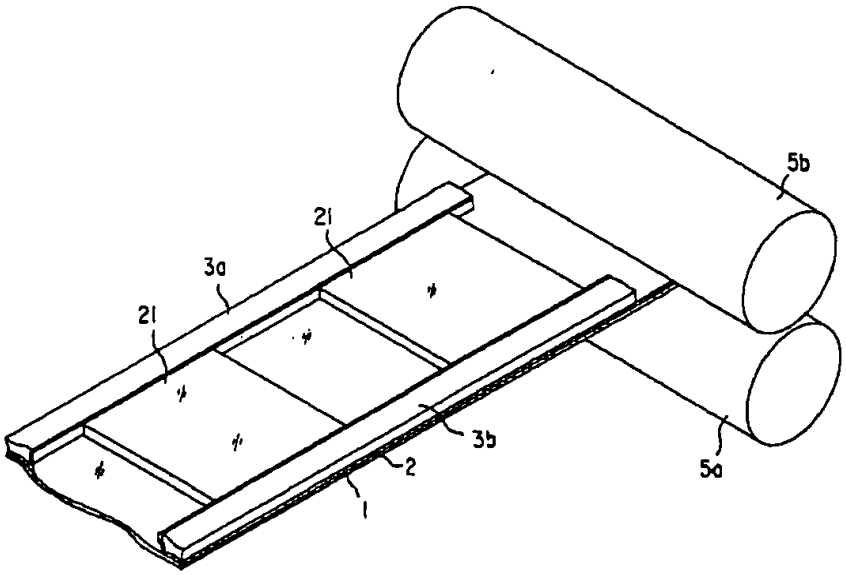
【図４】 [Figure 4]



【図３】 [Figure 3]

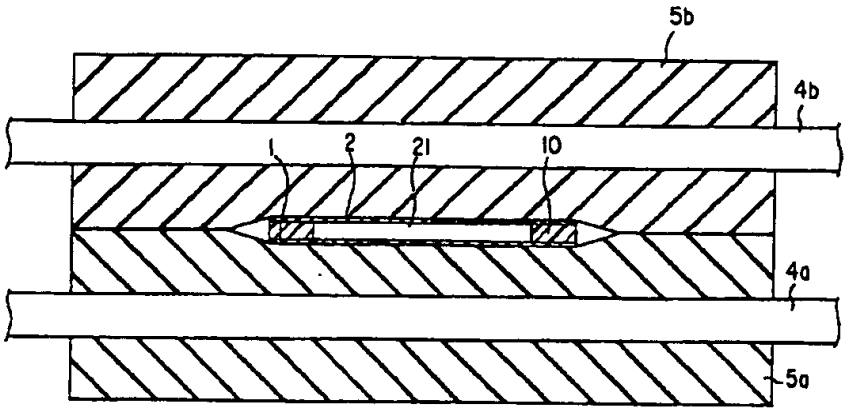


【図５】 [Figure 5]



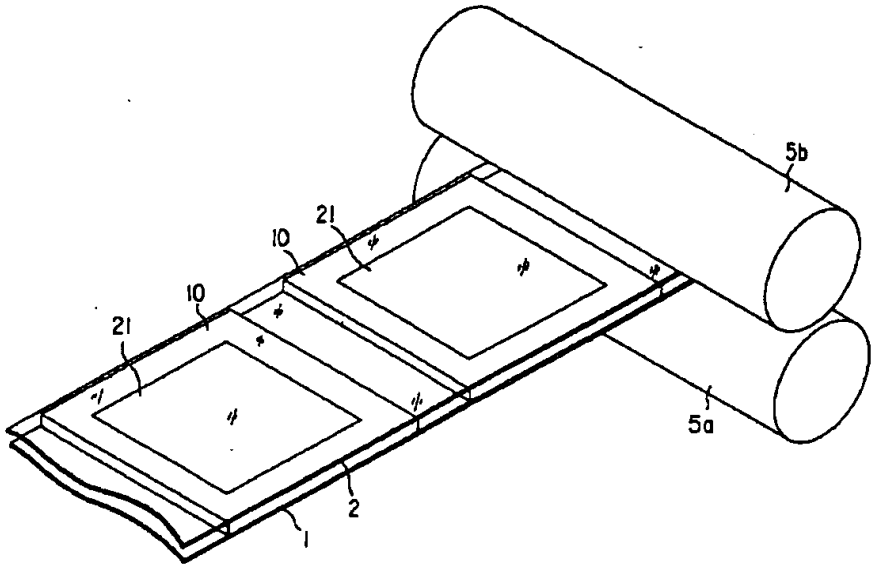
【図7】

[Figure 7]



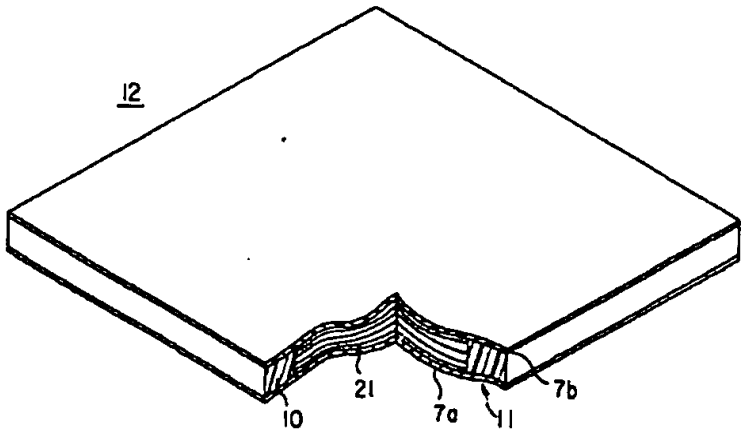
【図6】

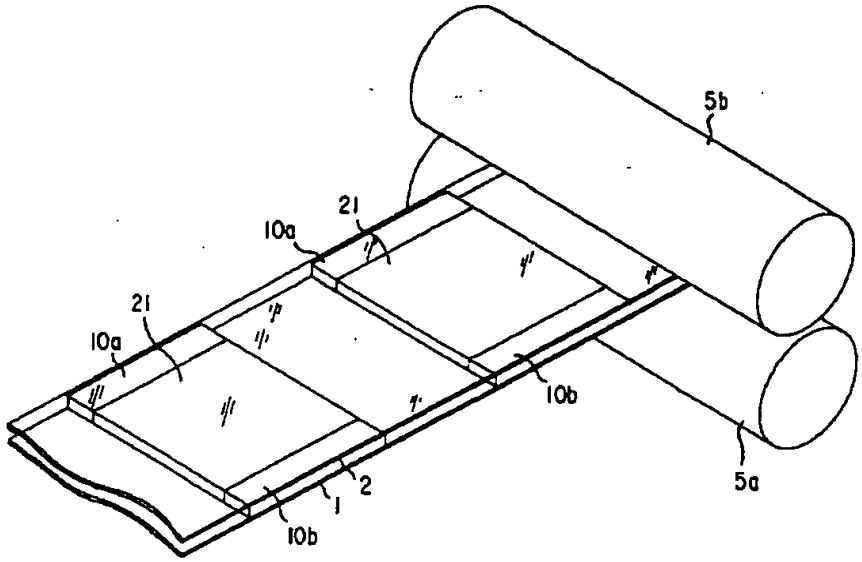
[Figure 6]



【図８】

[Figure 8]





【図９】

[Figure 9]